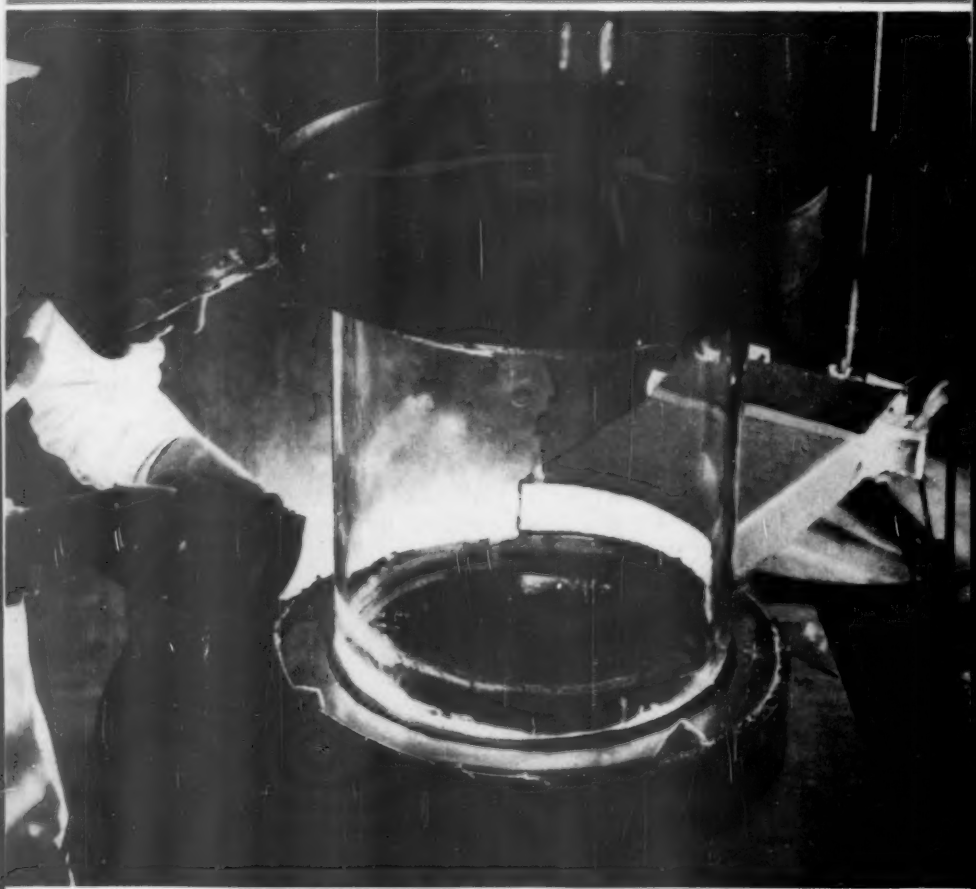
NUCLEONICS

TECHNIQUES AND APPLICATIONS OF NUCLEAR SCIENCE AND ENGINEERING

February, 1951



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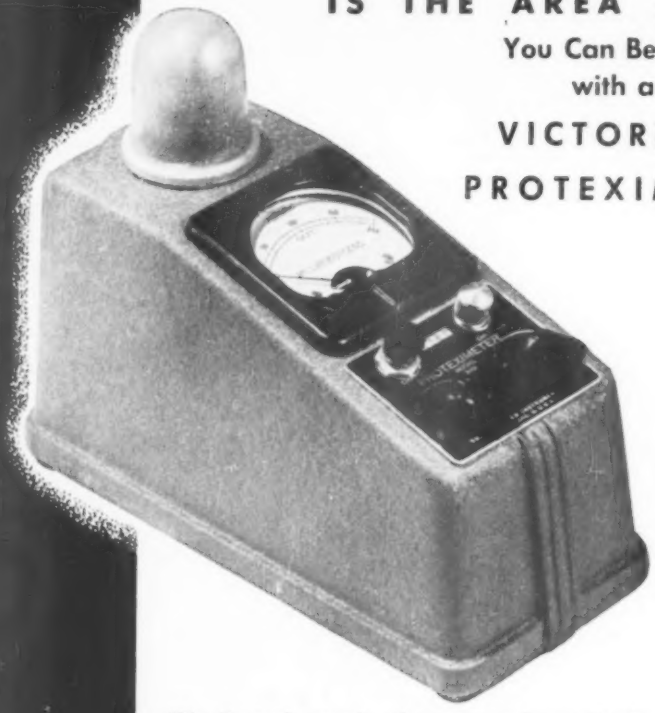

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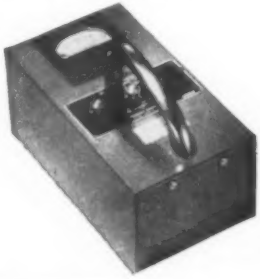

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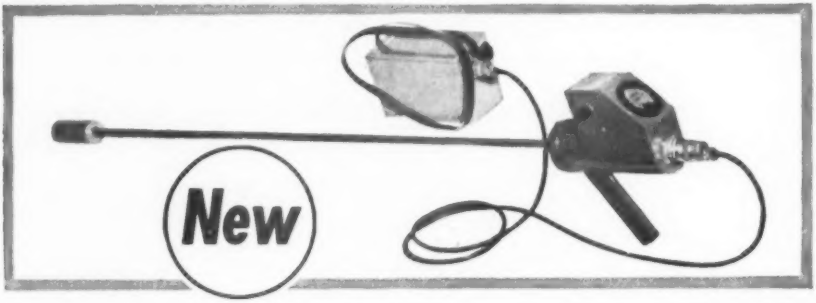

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Progress Report

TEN YEARS of ATOMIC ENERGY

"We have come a long way in the utilization of atomic energy since 1940, but as far as the development and use of nuclear reactors are concerned, we have only begun to scratch the surface."

This is the conclusion drawn from the panel discussion, sponsored by the Oak Ridge Institute of Nuclear Studies and the Oak Ridge National Laboratory, held in Cleveland, Ohio, on December 28, 1950, during the 117th meeting of the American Association for the Advancement of Science. The panel members were Dr. Walter H. Zinn, director of Argonne National Laboratory, Dr. Miles C. Leverett, technical director of the NEPA project at Oak Ridge, Dr. C. G. Suits, and Philip Sporn. The talks by Dr. Suits and Mr. Sporn centered on the economic aspects of nuclear power. Both of these talks appear on the following pages.

In his talk, Dr. Zinn traced the historical development of reactors, from the first one completed in 1942 to the more advanced ones presently being worked on. He also commented on the building of economically feasible nuclear reactors. Dr. Zinn said, "There is no dearth of ideas on how to build an economically feasible reactor." It is of interest to note here that Dr. Eugene P. Wigner, who was director of research and development at Clinton Laboratories during World War II and is now professor of physics at Princeton University, made the statement from the floor that enough information was available at the end of 1950, apart from the cost of raw materials, for satisfactory economic studies of nuclear power to be made.

For economically feasible nuclear power, Dr. Zinn stressed these points:

1. Reactors must be designed to be economically feasible; if a reactor is designed for military use, costs are not of primary importance.
2. Reactors must be designed for power levels of at least 100,000 kw.
3. It must not be expected that the first power reactor built will be competitive with other forms of power.

Another point which Dr. Zinn made is that reactors should not be multipurpose, but rather should be designed for only one purpose.

In his talk, Dr. Leverett pointed out that the present emphasis on military reactors may actually accelerate the development of reactors for power. He cited examples of how military developments had in the past aided peacetime progress.

Power from the Atom—An Appraisal

By C. G. SUITS

Vice President and Director of Research, General Electric Company
Schenectady, New York

THE DEVELOPMENT of useful power from atomic fission has been in progress in this country for about five years. As yet, no useful power of any consequence has come from the very considerable effort in this direction. For this and other reasons it is clear that this is a long term project, and the general realization of this fact represents progress.

The *next* stage of progress is the one

which we are now entering. In this second stage we can achieve, as a result of the excellent scientific and technical work of many laboratories, the beginnings of a real perspective on the problem of power from the atom. It has taken many years of research on the reactor problem to provide the solid foundation upon which such a perspective may be supported.

In addition to the laboratory work on power-producing reactors, there has been a vast accompaniment of public debate, discussion and comment on this subject. Perhaps never before has so much been said by so many with so few facts. What atomic power is, or is not—what it will do, or fail to do—every possible sociological and economic aspect of the matter—has been so thoroughly considered that little remains to be said. There is, of course, the necessity of obtaining the scientific facts upon which the whole conjectural structure rests; this will take another ten years.

It is safe to say, however, that atomic power is *not* the means by which man will for the first time emancipate himself economically, whatever that may mean; or forever throw off his mantle of toil, whatever that may mean. Loud guffaws could be heard from some of the laboratories working on this problem if anyone should in an unfortunate moment refer to the atom as the means of throwing off man's mantle of toil. It certainly is not that!

What then is atomic power; where does it fit into the picture? In short, what is all the shouting about? At present, atomic power presents an exceptionally costly and inconvenient means of obtaining energy which can be extracted much more economically from conventional fuels. If the economics of heat production were the only consideration, the great outlays for reactor research and development could not be justified. We have no shortage of fuel in this country. Whatever the elastic life span of petroleum may be, coal is certainly available for hundreds of years in the future. The economics of atomic power are not attractive at present, nor are they likely to be for a long time in the future. This is expensive power, not cheap power as the public has been led to believe. The development of nuclear reactors for power production *is*, however, very important for quite a different set of reasons. These reasons are:

1. Atomic energy is an extremely concentrated source which presents important military advantages in a mobile power plant.

2. Atomic plants may simultaneously produce large blocks of power *and* fissionable material needed for military purposes.

3. The scientific and engineering knowledge gained from intensive effort on (1) and (2) above may in the longer term make possible the building of economical plants primarily for the production of power.

These reasons are compelling and important and are strongly accented by the troubled state of the world. They are quite different from the reasons which seemed important in 1945.

Let us now examine some of the numbers which bracket the economics of atomic power plants. Although the basic information required for a precise appraisal of these matters is not available, there are several hypothetical cases worth considering, each of which corresponds to a set of technical possibilities and hopes. As a base line to which these hypothetical cases may be referred, consider the set of figures of Table 1 which gives for a typical steam-electric plant* of 400×10^6 watts (or 400 MW) electrical output, (a) the esti-

* See for example, Federal Power Commission Report S-83 "Steam Electric Plant Construction Cost and Annual Production Expense, 1949."

TABLE 1
Typical Steam-Electric Power Generating Plant—1950

Net station electric power output	400 MW
Auxiliary power requirements	37 MW
Gross output	437 MW
Net station heat rate	9,800 BTU/kwh
Gross heat input (400,000 × 9,800)	$3,920 \times 10^6$ BTU/hr
Gross heat requirement (maximum boiler capacity)	1,150 MW
Estimated construction cost	$40,000,000
Unit cost (on 400 MW net available)	$100/kw

Construction Cost Estimate

Site	1.%	= $ 400,000
Coal handling Ash handling	6.	= 2,400,000
Boiler plant	46.	= 18,400,000
Generator plant	40.	= 16,000,000
Switchgear, transformer	7.	= 2,800,000
	100 %	= $40,000,000

Estimated Electric Power Production Costs (at 70% utilization)

Taxes (at 2%) Interest (at 6%) Depreciation (at 3%)	1.8 mills/kwh
Fuel	2.
Operation	1.
	4.8 mills/kwh

mated efficiency, load utilization and plant cost, (b) the estimated construction cost breakdown and (c) the operating costs. Such figures have a considerable variation between different plants at different locations, so that the term "typical" must be used with some reservations.

For example, the operating costs per kwh (exclusive of fuel) for steam electric plants show an extreme variation of some 20 fold throughout the country, while the fuel costs per kwh also show a 20-fold variation. The reasons for this involve a great variety of practical considerations, including the age of the plant, operating efficiency, load utilization, size, location relative to centers of population, and source and kind of fuel and so forth. The figures of Table 1 have been selected to represent some of the desiderata of importance in the present discussion. An efficient atomic plant must be large—the steam electric plant of Table 1 is large. The location is presumed to be on the eastern seaboard where the quoted costs are believed to be realistic for late 1950. A different set of assumptions could be made with corresponding changes in the figures; the numbers of Table 1 will serve our present purpose, however.

Referring now to Table 1, we may see that the gross heat requirement is 1,150 MW, which could be supplied presumably by one or more nuclear reactors. The construction cost of the entire plant is at the rate of $100/kw. Under somewhat altered circumstances this might be $140/kw or even higher. Please note that the boiler plant, which is the unit for which we would substitute a nuclear reactor, is 46% of total plant cost, corresponding to $46/kw of the net electric generating capacity, and that the cost of equipment for handling coal and ash is only 6% of the total. Under the items of generating cost, note that fuel is 2 mills/kwh, operation 1 mill/kwh and that taxes, interest and depreciation amount to 1.8 mills/kwh.

The depreciation is figured on the basis of 3%/yr which is well supported by utility experience. The rate of depreciation of any power-producing reactor would have to be much higher; in the absence of experience I have assumed 10%/yr in the discussion below.

Case A

Now refer to Table 2 and especially the assumptions on which it is based. Here it is assumed that the cost of nuclear fuel on a heat basis equals conventional fuel. This might be realized in a variety of circumstances which would add up to the same result. The

TABLE 2

Hypothetical Nuclear Steam-Electric Plant of 400 MW Power Output in 19—

Case A

ASSUMPTIONS: 1. Nuclear fuel cost (including handling and processing) equals conventional fuel costs on a heat basis.
2. Operation costs the same.
3. No credit for by-products.

Credit from coal fired boiler plant	—	0.8 mills/kwh
Annual " " " " "	—	$ 2,000,000
Additional cost* nuclear boiler†	—	$11,100,000
Gross plant cost	—	$32,700,000

Case B

ASSUMPTIONS: 1. Nuclear fuel costs negligible.
2. Operation costs the same.
3. No credit for by-products.

Credit from fuel savings	—	2 mills/kwh
" " boiler plant	—	0.8 " "
Additional cost* nuclear boiler†	—	$38,300,000
Gross plant cost	—	$59,900,000

Case C‡

ASSUMPTIONS: 1. Nuclear fuel costs negligible.
2. Operation costs the same.
3. Plutonium credited at the rate of 2 mills/kwh.

Credit for fuel savings	—	2 mills/kwh
" " boiler plant savings	—	0.8 " "
" " plutonium	—	2 " "
Additional cost* nuclear boiler and chemical plant†	—	$65,500,000
Gross plant cost	—	$87,100,000

* With interest on investment at 6%, local taxes 2%, depreciation 10%.
† To produce electric power at same net cost as a conventional plant.
‡ For a plutonium plant.

weight comparison between fissionable fuel and coal for the same heat release is $1:3 \times 10^6$. The ratio of $1:10^6$ is about the relation between a gram and a ton. Hence the cost of fissionable fuel per gram may be about three times that of coal per ton. Reliable figures are of course not available, although it was stated by Dr. Hafstad* of the AEC in reference to a quoted figure of $20/gram that "The $20 figure is almost certainly low and seems likely to increase as the supply of high grade uranium runs out." Since the cost of handling fissionable materials and especially radio-ash will be high, it is likely that the cost of producing separated fissionable materials must be brought down remarkably if we are to use them in enriched power reactors.

It is altogether probable that these materials will be produced more cheaply in the future. G. R. Prout, General Electric manager of the Hanford Works, announced recently† that the cost of plutonium had been cut in half during the last four years. A considerable reduction in the cost of enriched fuel would help to offset processing costs which at present are very high.

* L. R. Hafstad, Director, Division of Reactor Development, U. S. Atomic Energy Commission—remarks before American Petroleum Institute, Los Angeles, Calif., Nov. 15, 1950.

† *New York Times*, December 4, 1950, p. 13

It is assumed in Case A that plant operation for the nuclear steam-electric plant is the same as for the conventional steam-electric installation. The physicist-chemist-encrusted gear which surrounds present reactors does not encourage this hope, but we have much to learn and must expect progress in this area. In the conventional plant a great volume of coal must be handled, but this is largely a mechanized operation. The nuclear plant, on the other hand, may under the best of circumstances handle a minuscule volume of fuel and one may expect that this operation will be highly mechanized in the future.

It is further assumed in this particular case that nuclear by-products are not worth reclaiming; or if reclaimed, the cost of processing offsets their value.

Under the stated assumptions, the higher depreciation rate on the nuclear boiler means that less capital cost can be tolerated for the same net electric power cost. Accordingly, the reactor costs for the 1,150 MW unit must not exceed approximately $11,000,000.

Case B

Similar hypothetical plants are discussed as Case B and Case C. In the former, the assumptions are the same, except the cost of nuclear fuel per heat unit is assumed to be negligible. This might be the case for example in a reactor in which native uranium would be "burned" until exhausted and then thrown away; or it might be realized in a nuclear economy in which the spent power reactor fuel could be sold for a by-product value approximately equal to its original cost.

In this case, the calculation takes account of the saving in fuel cost and of the omission of the conventional boiler. Credits from these items are allocated against the nuclear boiler, bearing in mind that it is depreciated at the rate of 10% per year, whereas the conventional boiler is depreciated at the rate of 3% per year.

Under these circumstances one might allocate a capital investment of $38,000,000 to the nuclear boiler of 1,150 MW heat output.

Case C

This case includes a combination of favorable circumstances which might develop in the long-term evolution of atomic power in a plutonium economy. The new assumption here is that an efficient nuclear fuel breeding cycle is established and that the "spent" nuclear fuel is sold for its plutonium content and credited against the power production at a rate of 2 mills/kwh. It is obvious that with a substantial by-product credit, a considerably greater investment in reactors can be justified. Under these assumptions, the reactor-boiler plutonium producer might cost $65,000,000 and still permit competitive costs on power production.

Reactor Costs

It would be interesting now to take a look at reactor costs for the purpose of determining whether their present magnitude or future downward extrapolation is likely to fall within any of the hypothetical brackets which have been cited.

The cost figures are collected in Table 3 for convenience. Since the cost of most reactors has not been made public,

TABLE 3
Boiler Plant Estimated Costs
1,150 MW Heat Output

Conventional boiler	$18,400,000
Nuclear boiler (Case A, fuel and operation cost equal to conventional)	11,100,000
Nuclear boiler (Case B, fuel cost negligible)	38,300,000
Nuclear boiler (Case C, by-product credit)	65,500,000

and since no power-producing reactors have been built, and since the ones now being built will be much smaller than what will be finally required, a great deal of guessing must be done from this point on—few really pertinent facts are available.

I believe the only public information available* on a reactor for which both power level and cost are known is the experimental reactor at the Brookhaven National Laboratory. This unit, rated at 30 MW, is stated to cost approximately $25,000,000.

Extrapolating to high-power reactors from the baseline provided by the Brookhaven reactor involves many uncertainties. This reactor is primarily an experimental unit which produces only a token amount of power incidental to its cooling. Operation at the high temperatures required to produce power efficiently would necessitate a change in design and probably a higher cost for the same power. On the other hand, the provision for experimental facilities inevitably adds greatly to the cost of any reactor. More important than either of these considerations is the fact that these small experimental reactors properly include factors of safety with regard to their thermal and nuclear design, their controls, their materials of construction, and provisions for flexibility of operation and inspection which will be considered unnecessary and excessive in practical power reactors.

The mere extrapolation of a reactor from pilot plant size (10 MW–30 MW) to practical power ratings (100 MW–1,000 MW) permits substantial cost savings per unit of output. Although a reactor for higher power must generally be larger, the increase in size will be much less than the increase in power. As a broad generalization of some validity, power reactors of a given basic design tend to have a more or less constant size over a considerable range of power output. The heat transfer system must increase in size and cost as power increases, but the weight and cost of the shielding, which is a very substantial item of cost, does not change much.

It is evident from a scrutiny of the materials of construction now available and from the factors of safety which we must employ that we are paying a high price for the elementary state of our knowledge of reactor technology. A substantial factor of improvement must be expected from continuing research and development in this area.

After first considering and weighing all of these factors, one may estimate that the cost of building a power production reactor of 1,000 MW output under current limitations of technology might be in the range of $50,000,000 to $100,000,000.

Conclusions

It is rather surprising to find that there is not a greater divergence between economically permitted reactor costs and the postulated costs of reactor construction. Although it should be emphasized that the underlying assumption must be borne in mind, it should also be pointed out that the economic baseline presented by Table 1 does not by any means represent the most favorable set of circumstances for reactor economics. A combination of high fuel and transportation costs coupled to the requirement for large blocks of power would considerably enhance the case for nuclear power plants, and it is probable that such cases will provide initial opportunities for exploiting this new power source.

The future solution of difficult technical problems will be accomplished partly with knowledge now at hand, and partly with new knowledge which we are certain to gain from continuing

* AEC Release #219, Nov. 28, 1949; see NUCLEONICS **6**, No. 1, 94 (1950).

scientific research. On a long-term project it would be a great mistake to underestimate the importance of the latter to the final solution.

Development of Atomic Energy— How Can Private Industry Best Participate?

By PHILIP SPORN
President, American Gas and Electric Service Corporation
New York, New York

IN THE SPHERE of economics, it is no secret that we have made little headway in reactor development. No nuclear reactors are supplying any part of our power requirements, and there is no clear indication as to when they will commence to do so. I do not mean to say that this is a good thing, but there is no use quarreling with the facts as they are. On the contrary, I propose to ask, and I think it will be useful to do that, why this is so.

A formidable array of technical obstacles stands in the way of getting ahead. It is with these problems that the National Laboratories have been preoccupied since 1945. Progress, perhaps much progress, has been made toward solutions, but until we reach the stage of pilot plant trials, it will be impossible to assess the exact state of the art.

But the technical difficulties in the way of perfecting economical power-producing reactors are only a part of the problem.

Among the deep reasons why we are not farther ahead is the fact that, relatively speaking, the state of power production and development by conventional means, particularly in this country, is unusually satisfactory. Thus, in the United States, with less than 7% of the world's population, we now produce more than 40% of the world's electricity. In the period 1949 through 1952 we will have completed installation of new electric facilities to increase our total supply by close to 29,000,000 kw—more than 50% of the capacity installed at the end of 1948. The ability to produce electricity and the facilities for making more electricity in such relative abundance does not make for stimulating heroic efforts to develop radical new means of production.

Moreover the efficiency of conversion in our most efficient steam plants has risen from 13 to 15% just a few years ago to 38% thermal efficiency—9,000 BTU per kwh—in the best plants now being brought in.

One might almost say that one of the obstacles to rapid progress with nuclear power production is the rather bright prospect for conventional power production.

We keep overlooking the fact, too, that nuclear power is not a new kind of power but is only a *new fuel*. As such,

EDITOR'S NOTE: *The remarks of Mr. Sporn are of particular interest at this time because of his position as chairman of the Advisory Committee on Cooperation Between the Electric Power Industry and the Atomic Energy Commission. With a number of proposals by industrial organizations before the AEC at present, it is expected that the Sporn Committee report will have considerable influence on the decisions of the AEC in these matters.*

it will be used in processes that have been under development ever since the steam engines of Newcomen and Watt. So, even after all technical obstacles to perfecting a power reactor have been overcome, the most that we can hope for on costs is the cheapening of one of the elements in power production, namely, fuel. But, while this happens we must also expect some—perhaps a large—increase in the capital cost of certain elements of the plant. And there does not seem to be any doubt that operation and maintenance of a nuclear reactor will be more expensive than the corresponding part in a conventional plant, the thermal reactor.

Even so, the prospect of a radical reduction in the cost of the fuel component for power production and the hope that this can be accomplished without an increase in capital cost so great as to render nuclear reactors economically unsound is a sufficient reason to make industry vitally interested in the development of reactors.

Nor is there any inconsistency in this frank statement that while industry is —and should be—vitally interested in the development of nuclear reactors, there has been up to now an absence of sufficient challenge to industry in this field to produce a fully adequate response. The absence of challenge, moreover, has been coupled with a lack of opportunity—a point which I shall come to a little later. But for the moment I should like to examine the question of industry's interest in atomic power.

Importance of Atomic Power to Industry

The problem of bringing about practical—and here I give special emphasis to the economic phase—atomic energy or power is admittedly difficult. Why, one might ask, should one persist? What makes it so important not to give up trying to bring about solutions to these difficult problems?

Development of nuclear power is important to industry not because of expected revolutionary consequences. It is important rather because it would provide a more universally available, more readily portable, and more economical fuel. For, if the more optimistic ideas as to breeding possibilities are realized, the fuel cost would be reduced to almost zero.

Chester F. Smith has recently presented* the results of a study of the part of the energy market supplied by four sources: hydro power, natural gas, coal, and oil, over the past decade and the expected relationships during the coming decade. These are as follows:

Sources of Energy Used in the U.S. (10^{15} BTU)

Year	*Hydro Power*	*Natural Gas*	*Coal*	*Oil*	*Total*
1940	0.9	2.8	12.7	7.8	24.2
1949	1.6	5.8	12.2	12.4	32.0
1960 (est)	2.3	9.8	13.6	17.2	42.9

With hydro bound to decrease, as better sites become more and more used up, it is apparent that if *another highly economical fuel* gave promise of having possibilities for development, natural pressures for such development will build up, if not before then, then surely when the reserves of present highly economical supplies, both in oil and gas, begin tapering off.

Such development will have an important bearing on those industries using fuel directly and on those—by far the larger percentage—using fuel as a source of electric energy. For it will not only protect and keep whole the entire base of economical fuel supply, but it might also lead to further expansion of use of electricity. For if cheaper power can be achieved through the use

* "Opportunities for Petroleum in the Energy Markets of the Future," 30th Annual Meeting of the American Petroleum Institute, Los Angeles, Calif., Nov. 15, 1950.

of atomic energy, several industries might well be stimulated. Examples are: reduction of magnesium and aluminum, refining of copper, production of cement, chlorine and caustic soda, electric furnace operations like the production of graphite, carborundum, calcium carbide, phosphoric acid, ferro-alloys and electric steel. All these operations are characterized by a high figure of kilowatt hours per week per worker and a relatively low figure of wages per kilowatt hour used.

To put it another way, power costs in these industries represent a substantial percentage of total cost—as contrasted with a relatively minor percentage in most industries—so that an appreciable reduction in power costs, as by lower fuel cost, will materially reduce cost of product. The resultant application or use is bound to be stimulated.

The industries, then, that could benefit most from any appreciable reduction in the cost of electric energy are those just enumerated. The displacement of other forms of energy by atomic energy in these industries will be indirect. Atomic fuel will displace coal or gas or oil in new plants, or in rebuilt old plants, or white coal in hydro plants that might otherwise be built.

Some new electric processes might also be developed if atomic power is cheap enough: electric melting of general-purpose glass, for example; or direct reduction of iron by electric processes; certain kiln operations might be electrified.

Whether or not atomic power will stimulate these existing processes or encourage the development of such new ones will depend on cost.

Participation of Private Industry

How can private industry best participate in the development of atomic energy? This question must be approached against the background just sketched: tough technical problems in the solution of which the traditional role of industry is more than ever necessary; no spur of pressing economic necessity to drive industry into atomic energy work because conventional power supply is at least good and its future prospects are also bright. But at the same time there is substantial incentive to proceed with atomic power development because, if successful, it promises not inconsiderable advantages in abundance, cost and convenience of fuel supply.

Now thus far private industry has not had much part in atomic energy development. This has been only partly because economic advantages of successful nuclear power appear moderate rather than very high. There is already considerable history on the subject of why industry does not have a genuinely important role in atomic work. The report of the AEC Industrial Advisory Group (NU, Feb. '49, p. 77) over two years ago gave an excellent analysis of this question and made a significant beginning in suggesting the means by which industry could be effectively brought into atomic energy.

The Industrial Advisory Group report, while commending the AEC contract method of operation, demonstrated that that method represented only limited participation as compared with what was required if industry was to make a real contribution. As the report so succinctly states:

> "The difficulty and the danger in the present situation is that industry's part in atomic energy is very limited as compared with the opportunities which exist and always have existed in other fields. The small number of companies which take significant part are selected by the Government and the extent of their role in the work is limited by specific assignments from the Government."

Probing for the cause, the Industrial Advisory Group further states:

> "We think that today the central difficulty in getting a broad industrial

attack on the problems of atomic energy is the fact that industry has no way of determining whether important opportunities in fact exist in which to take part."

And again:

"It has been stated that industrial opportunities in atomic energy are potentially unlimited. But they are at present so shadowy that businessmen neither know where to look nor what to look for. Today no one can say whether the prospect of profits or other incentives exist, because under present conditions the great majority in industry know little or nothing about the subject.

"The need for Government monopoly in certain important areas, coupled with secrecy, seems to erect an impenetrable barrier to a wish for knowledge. If industry is to help in atomic energy and benefit from it, industrialists must first be put in a position to find out enough about the subject to determine whether and where there are in fact opportunities to take part. In devising means to this end, only the Government can take the initiative. The Government, which must exercise a broad monopoly and determine security regulations, alone is in a position to open the doors. Industry can, and we are confident that it will, cooperate. But Government must first provide the catalytic forces that will set more of the normal processes of industry to work."

Even though more than two years have elapsed since the submission of this report, the basic difficulty so clearly pointed out by the Industrial Advisory Group has not been much alleviated. Industry still needs the things the Industrial Advisory Group advocated: More information about atomic energy. More opportunity for direct personal contacts between individual industrialists, technicians and executives, and the atomic energy program. Greater utilization by government of the Industrial Advisory Group concept.

The necessity for these things is clearly apparent today in any critical examination of the causes of the failure of private industry to participate or become involved to a greater extent than it has in atomic energy.

We are still suffering from atomic illiteracy.

There is a very deadly parallel between the present situation with regard to atomic illiteracy and the situation that existed shortly after World War I in the field of electronics. The electronic art made great strides, mostly through the development of military applications in World War I, but when the war was over what we had so far as the civilian population was concerned, or at least that part of it that didn't participate in the military program, was a state of almost complete electronic illiteracy. The consequence of that was that the predicted progress made during the early 20's did not fully materialize. It took a generation almost for that condition to cure itself, and it wasn't until World War II, more than a quarter of a century later, that the full fruition of what was started electronically in World War I took place.

The situation with regard to atomic energy in the post World War II period is much worse. Except for the relatively few people who are now engaged in the various laboratories and those among the various manufacturing organizations who participated and are participating in the atomic energy program, an almost complete state of illiteracy exists in the atomic field today in the United States. A few of our college professors and graduate physics students have some knowledge of the subject, the people of the National Laboratories possess it, and a limited number of people among the major manufacturers also. But, considering the size of the technical population of the United States, not to mention what it is if you consider the size of the business population, this group is woefully small.

Generally speaking, there is still no

(Continued on page 104)

Estimating Radioelements in Exposed Individuals—I*

RADIOELEMENT METABOLISM

With an increasing number of workers handling radioactive materials, more data are needed on the fate of radiation in the body. Based on considerable research covering the physical chemistry and metabolic behavior of certain radioelements, this series of papers presents a thorough discussion of the theoretical and practical considerations involved

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INDIVIDUALS who work with or near radioactive substances may, through accident or carelessness, accumulate appreciable levels of radioelements within the body. By periodic radiochemical analyses of excreta, it is possible to estimate the amount of a given radioelement fixed in the body at a given time.

The physical aspects relating to radioactive hazards, namely, study of the interaction of radiation with the human body and prevention of radiation damage to personnel, have been treated at some length by Morgan (*1*, *2*), Parker (*3*), Evans (*4*), Ingram (*5*), and Failla (*6*, *7*). The Atomic Energy Commission has issued recently a comprehensive report on the "Control of Radiation Hazards in the Atomic Energy Program" (for sale by the Superintendent of Documents, U. S. Government Printing Office, Washington 25, D. C., 55 cents).

The metabolism of some radioelements is largely independent of factors such as the physiological state and yet greatly influenced by the route of administration. The reverse situation holds for other groups of radioelements. However, it is possible to anticipate the relative importance of chemical and physiological conditions on the distribution and metabolism of a given radioelement by considering some aspects of the solution chemistry of cationic radioelements, particularly under physiological conditions.

In addition, the study of the interactions of radioelements with synthetic organic cation exchangers can be used to correlate a large amount of metabolic data and permit predictions to be made regarding the effects of various factors on the normal metabolic patterns.

Aspects of the Solution Chemistry of Cationic Radioelements

A useful index for anticipating the metabolic behavior of a cation is the *p*H at which the hydroxide of a dilute solution of a simple salt of the element will precipitate. A tabulation of such values is given in Table 1. The recorded *p*H values actually are maximum values, since colloidal solutions are formed approximately a *p*H unit before observable precipitation. This is reasonable in view of the fact "that the

* Part II, on the subject of radiation dosage and permissible levels, will be published in the March NUCLEONICS; Part III, describing bioassay operations and procedures, will appear in the April NUCLEONICS. All three parts of this paper are published on the sole responsibility of the author. The recommendations offered do not represent the official views of any laboratory or division supported by the U. S. Atomic Energy Commission.

process of precipitation is one which involves the formation of a colloidal solution which subsequently coagulates" (p. 59 of ref. *8*).

The development of colloidal properties by a solution cation is associated with a tendency to polymerize as a result of repeated hydroxylations. This results in the formation of particles whose diameters exceed 10 A.U. (10^{-7} cm). At a *p*H of about 7.2, most of the elements listed in Table 1 exhibit some colloidal properties, namely, slow diffusion rates, either free or through membranes; a tendency to undergo irreversible adsorption on surfaces; discontinuous distribution of radioactive elements as revealed by radioautographs; incomplete exchange or lack of equivalence between radioactive and nonradioactive isotopes of the same element; and anomalous adsorption by ion exchangers. These colloidal properties are observed even with tracer levels of radioelements—the so-called radiocolloids (*9*). The tendency to become colloidal, from the standpoint of rate, extent, and degree of irreversibility, increases the lesser the *p*H at which a solution of simple salt will precipitate.*

Chemical factors other than *p*H will induce colloidal behavior. These include combinations with anions, such as phosphate to form salts of low solubility, and combinations with proteins.

The presence of organic acid anions, such as citrate, often prevents or minimizes the formation of colloids by combination with the cation to form a soluble complex ion. An important point to consider for therapeutic purposes is that once a hydrolyzable element precipitates it often requires large unphysiological amounts of a complex-forming agent to redissolve it.

Mechanisms for Interpreting Metabolism

Ion exchange and colloidal adsorption are the mechanisms for consideration in interpreting the metabolism of radioelements, particularly bone deposition.

Cation Exchange Reactions: Consider first the reactions between cations and synthetic cation exchangers, such as the Amberlites and Dowex resins (*12, 13*). The cation exchanger will be assumed to be in the BR form, where B^+ represents a monovalent cation, and R represents the insoluble anionic part of the cation exchanger. When BR is immersed in a salt solution of B^+ containing tracer levels of the radioactive monovalent cation A^*, the cation exchange reaction is expressed by:

$$A^* + BR \rightleftharpoons A^*R + B^+ \qquad (1)$$

The exchange constant $K_{ex.}$ for this reaction, neglecting activity coefficients, follows from the law of mass action:

$$K_{ex.} = \frac{(A^*R)(B^+)}{(A^*)(BR)} \qquad (2)$$

where the quantities (B^+) and (A^*) represent molar concentrations and (A^*R) and (BR) represent mole fractions. For the conditions where (B^+) and $(BR) \gg (A^*)$ and (A^*R), the ratio $B^+/(BR)$ is a constant. The quantity (A^*R) is expressed as (%A^* in exchanger)/(mass of exchanger), while (A^*) is equal to (%A^* in liquid phase)/(volume of liquid phase). Under the stated conditions, we have:

$$K_{ex.} = \frac{\%A^* \text{ in exchanger}}{\%A^* \text{ in liquid phase}} \times \frac{\text{volume of liquid phase}}{\text{mass of exchanger}} \times C \qquad (3)$$

---

* It is probably more exact to relate the colloidal tendencies to the acid or hydrolysis constant, K, defined as:

$$K = \frac{(H_3O^+)[R(OH)]^{+(n-1)}}{(R^{+n})}$$

which is the mass action equilibrium expression for the reaction

$$R^{+n} + 2H_2O \rightleftharpoons R(OH)^{+(n-1)} + H_3O^+$$

Depending on the basicity of the cation R^{+n}, the process continues to yield a series of acid constants. For example,

$$R(OH)^{+(n-1)} + 2H_2O \rightleftharpoons R(OH)_2^{+(n-2)} + H_3O^+$$

As the *p*H increases the reactions continue to a point where a colloidal solution is formed, presumably because of polymerization of the hydroxylated compound which occurs through the binding of various number of atoms of the cations by R-OR linkages and with varying numbers of hydroxyl groups attached (*10, 11*).

TABLE 1

pH at Which Precipitation of a Hydroxide or Basic Salt Will Take Place from Dilute Solutions of Simple Soluble Salts of Cations*

Element	pH	*Element*	pH	*Element*	pH
Magnesium	10.5	Cobalt	6.8	Aluminum	4.1
Silver	7.5–8.0	Yttrium	6.8	Mercurous	3
Manganous	8.5–8.8	Cadmium	6.7	Thorium	3.5
Lanthanum	8.4	Nickel	6.7	Indium	3.4
Cerous	7.4	Lead	6.0	Ceric	2.7
Mercuric (chloride)	7.3	Beryllium	5.7	Plutonium (IV)	2.3†
Praseodymium	7.1	Ferrous	5.5	Mercuric (nitrate)	2
Neodymium	7.0	Cupric	5.3	Stannous	2
Zinc	7.0	Chromium	5.3	Zirconium	2
Samarium	6.8	Uranic	4.2	Ferric	2

* After Britton (Vol. II, p. 79 of ref. *8*).
† Taken from data of Kraus (*61*).

where C is a constant equal to $(B^+)/(BR)$. This leads to a definition of the distribution coefficient, K_d, as:

$$K_d = \frac{\% \text{ of tracer in exchanger } (M_s)}{\% \text{ of tracer in liquid phase } (M_l)} \times \frac{\text{volume of liquid phase}}{\text{mass of exchanger}} = \frac{\lambda v}{m} \quad (4)$$

where λ represents the ratio M_s/M_l.

Changes in the composition of a solution system are reflected in the variation or constancy of K_d. In the case of a *reversible cation-exchange* reaction involving tracer levels of a radioelement at constant ionic strength and *p*H, a variation in v/m is offset by a corresponding change in λ, so that K_d remains constant. The effect on K_d of increasing the concentration of tracer can be calculated (p. 177 of ref. *13*) from an expression directly derivable from Eqs. 2 and 4:

$$K_d = k(\text{BR}) \quad (5)$$

where k is a proportionality constant and (BR) designates the concentration of B^+ in the exchanger. As long as $(BR) \gg (A^*R)$, no change in K_d can occur regardless of the extent to which (A^*) is taken up by the exchanger. At the point, however, where the concentration of (A^*) constitutes an appreciable fraction of the concentration of (BR), then a corresponding change in the per-cent uptake of (A^*), and hence, in K_d, follows as the value of (BR) diminishes. An example of the use of Eq. 5 will be shown in connection with the uptake of radiostrontium by bone.

Often a situation arises (*13*, *9*, *14*) in which the per-cent uptake of a radioelement by a cation exchanger remains practically unchanged despite large changes in the v/m ratio. Further, the per-cent uptake, and hence K_d, may even increase as the bulk or supporting electrolyte concentration in solution is increased. Often a notable characteristic of such anomalous uptake is the pronounced irreversibility of the reaction, i.e., the adsorbed tracer cannot be removed to any appreciable extent except by the use of substances which react readily with the tracer to form soluble complex ions. Adsorption of this kind, termed *colloidal adsorption*, occurs when the exchanger acts as an adsorbent for coagulated or charged colloidal particles.

Whether a cationic radioelement will be taken up by a reversible cation-exchange reaction or by colloidal adsorption can be gaged by the *p*H and composition of the medium. The condi-

Radioelements in Bone—

Radioelements are incorporated in bone by ion exchange, colloidal adsorption, or by mixed crystal formation. The distribution of the radioelement within the crystal is a function of several variables.

Consider a solution containing a tracer cation and from which calcium phosphate is precipitated. There are two ways in which the tracer can be incorporated by the calcium phosphate crystals—by isomorphous carrying or by surface adsorption. The latter case is discussed by Hahn (*15*). Isomorphous carrying is the mechanism we are most concerned with here. A true equilibrium is reached when all the crystals are in equilibrium with the liquid phases and the distribution of the tracer and the macrocomponent (calcium phosphate) in the solid is homogeneous. In such a case, the equilibrium constant follows from the mass action expression as given in Eq. 2 and is known as the Berthelot-Nernst distribution (Hahn). This situation requires a rapid migration of the tracer in the crystal or, in the case of very small crystals, a homogeneous distribution is obtained through *recrystallization*. In other cases the mass action Berthelot-Nernst equation may apply only at the instant that each increment of the precipitate is formed. Consequently the successive crystal layers will contain relatively less and less of the tracer because of depletion of the solution phase. The reverse situation also holds, i.e., the outside layer may be enriched at the expense of the inner layers. The occurrence of either case depends on the solubility product of the bulk components relative to the micro components.

The equation describing these situations is known as the Doerner-Hoskins equation (*16*). It can be derived from the Berthelot-Nernst equation by suitable integration. The equation is given by the expression

tions for judging whether a cationic radioelement is likely to be in a colloidal state were described in the discussion of solution chemistry.

The information in the two preceding sections will now be applied to metabolic data. To supplement these data, attention is directed to the information boxed above. It is a necessarily oversimplified physical-chemical picture of how radioactive substances are incorporated in bone. The purpose in presenting it is to direct attention to important *quantitative* concepts that have widespread implications for bone chemistry.

Ion Exchange Uptake by Bone: Bone can be considered to act as either a cation or anion exchanger containing a variety of functional groups. The capacity of bone for cation exchange can be expressed in terms of the total milliequivalents of *replaceable* calcium. About one fifth of bone calcium or 1 millimole per gram of bone is exchangeable (*22*).

On the basis of their chemical properties, the alkaline earths would be expected to undergo cation exchange with bone. The reaction of radiostrontium, Sr^{++}, is given by the expression (*23*):

$$CaA + Sr^{++} \rightleftharpoons SrA + Ca^{++} \quad (6)$$

where A represents a composite of all the functional anionic groups in bone.

Consider the effect of increasing the concentration of radiocalcium in an *in vitro* experiment where a given weight of ashed bone is equilibrated with a constant volume of solution. In the

—A Physical-chemical Picture

$$\ln \frac{a}{a-x} = \lambda \ln \frac{b}{b-y}$$

where λ is a constant, a and b are the initial concentration of the tracer and bulk component respectively, and a-x and b-y are the concentrations of each after precipitation. Factors such as the rate of formation of the crystals, and the degree of supersaturation affect the nature of incorporation of the tracer, and aging of the precipitate.

"Aging" of a precipitate involves such factors as a change in crystal structure or polymorphic transition, crystal perfection, agglomeration, cementing of particles, and Ostwald ripening (growth of large crystals at the expense of small ones) (*17*). An attempt to treat the kinetics of the uptake of isomorphous substances has been made by Imre (*18*) and others who relate the uptake as a series of first-order processes. Considerable work has been done on relating quantitative factors such as supersaturation, concentration, rate of crystal growth, and so on. Of particular note are the studies of Becker and Doring (*19*) and Volmer (*20*).

Some studies by Kuyper (*21*) are of interest in connection with the composition of calcium phosphate precipitated from salt solutions under physiological conditions of *p*H, temperature, and ionic strength. When calcium, magnesium, phosphate, bicarbonate, and citrate are present in the supernatant solution in the amounts in which they are present in serum ultrafiltrate, the precipitate contains these substances in approximately the same molecular ratios in which they are present in bone or dentine. When present in the supernatant solution in the amounts in which they are present in saliva, the precipitate contains them in roughly the same molecular ratios in which they are present in enamel.

experiments of Falkenheim *et al.* (*22*), 50 mg of bone ash were equilibrated with 25 ml volumes of solution containing tracer Ca^{++}. The bone sample presumably contained 0.05 mmole of exchangeable calcium. Based on Eq. 5, we have:

$$K_d = k(\mathrm{Ca}A) \qquad (7)$$

With tracer levels of Ca^{++}, about 75% of the tracer was adsorbed. With a solution less than 10^{-4} mole in Ca^{++}, i.e., less than 0.0025 mmole of tracer alkaline earth, no appreciable change in K_d is found since

$$0.05 - (0.75 \times 0.0025) = \sim 0.05.$$

However, a solution 10^{-3} mole or 0.025 mmole, if adsorbed to the same extent as tracer levels, markedly reduces the value of $(\mathrm{Ca}A)$, and hence a sharp drop in per-cent adsorption must take place at 10^{-3} mole, as was observed (*22*).

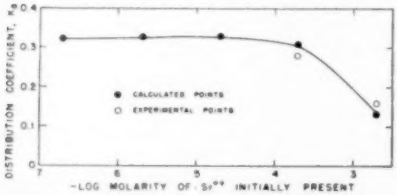

FIG. 1. Variation of K_d with strontium concentration for system in which powdered bone is equilibrated with $SrCl_2$ solutions (*24*)

In similar experiments (*24*) on Sr^{++}, adsorption dropped markedly at a concentration of $SrCl_2$ of 2×10^{-3} mole. If one assumes that 40% of the tracer was exchanged at tracer levels, then by Eq. 7 it can be calculated that a sharp drop in uptake should occur at 2×10^{-3} mole. See Fig. 1. The calculation

TABLE 2

Effect of Age on Content of Pu^{239} and $Sr^{89,90}$ in Liver, Kidneys, and Femurs of Mice Two Days after Intraperitoneal Injection

	32-day-old mice	*339-day-old mice*
% dose in femur		
Pu	2.02 ± 0.08*	2.28 ± 0.18
Sr	3.00 ± 0.12	1.53 ± 0.24
% dose in liver		
Pu	32.9 ± 3.1	36.3 ± 6.8
Sr	0.074 ± 0.001	0.085 ± 0.003
% dose in kidney		
Pu	1.42 ± 0.20	1.01†
Sr	0.020 ± 0.003	0.033†

* Each figure is the average obtained from two littermate mice unless otherwise indicated, and is followed by the average deviation.

† Data from one mouse discarded because of kidney disease.

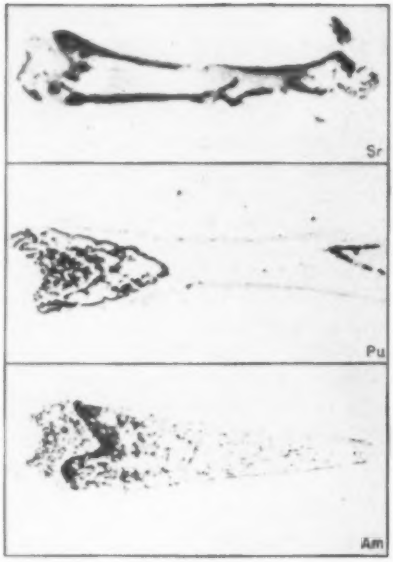

FIG. 2. Radioautographs of femurs of rats injected with strontium, plutonium, americium, respectively. [Hamilton (*46*)]

is as follows: at the tracer level, $K_d = 40/60 \times 25/50 = 0.33$. When 2×10^{-3} mole of Sr^{++} (0.05 mmole) is present, then $(CaA) = 0.050 - (0.4 \times 0.05) = 0.03$. Hence K_d to a first approximation should decrease to $0.33 \times [(0.05 - 0.03)/0.05] = 0.13$, which corresponds to an adsorption of 21% as compared with an observed value of 24%.

Similar calculations may be made for anionic uptake by phosphorus [exchangeable capacity for P = 0.06 mmole/gm of bone (*25*)] and fluoride. At very high concentrations of the tracer cation, the simplified Eq. 7 does not apply, but in all instances, one can place with marked accuracy that concentration of the tracer where K_d will undergo a sharp decrease. The main information required is the exchangeable capacity of the bone for either the cation or anion exchange process.

Analogous calculations can be applied for *in vivo* cases (*23*). Copp and Greenberg (*26*) injected nonradioactive "carrier" strontium to dilute and hence minimize deposition of radiostrontium. From Eq. 7, it can be calculated that about 60 mg of Sr would be needed in a 200-gm rat to reduce the bone deposition of radiostrontium (i.e., the point where a sharp drop in K_d would occur). This was found experimentally, but such an amount of Sr is toxic.

Age affects the uptake of the alkaline earths. The skeleton of a growing animal has a larger fraction of available sites for exchange than that of an adult. This is equivalent to increasing m in Eq. 4, in which case K_d, or the per-cent uptake by the skeleton, should be greater in young animals than in adults, all other factors being equal. This is shown in Table 2 from unpublished data of Schubert and White.

As expected from mass-action relations, the uptake of the alkaline earths is affected by changes in calcium level. However, in man, the amount of cal-

cium that can be removed from bone is too small in relation to the amount of exchangeable calcium present to reduce appreciably fixed deposits of radium, for example.

Colloidal Adsorption Uptake by Bone. From the precipitation values listed in Table 1, it is apparent that radioelements such as plutonium, zirconium, and thorium must become colloidal in the absence of high concentrations of a strong complex-forming agent at a *p*H of 7.2 following injection in animals. The bone uptake of such elements cannot take place by ion exchange because no simple ion forms exist.

Therefore, the uptake should have the characteristics of colloidal adsorption described earlier in terms of the variations of K_d. Consequently, the following properties should be associated with the bone metabolism of hydrolyzable radioelements:

1. The uptake by bone should be independent of the age or nutritional state of the animal. This follows from the fact that the per-cent adsorption is independent of changes in v/m.
2. The bone uptake should be highly irreversible, and the rate of elimination from bone extremely slow. No shift in position of the deposited material can be expected.
3. Treatments such as decalcification should be without effect since the uptake does not involve exchange with calcium.
4. Bone uptake would be on the surfaces of bone rather than throughout mineral structure.

Experimental confirmation of these expectations has been observed in numerous experiments (*27*, *28*). For example, experiments of Copp *et al.* (*27*) showed that calcium deficiency had no effect on the uptake of hydrolyzable elements Pu and Y in contrast to an alkaline earth, radiostrontium.

Radioautographs offer further support. No change in location of Pu is found in radioautographs taken at different times after injection. Furthermore, Pu and other colloidally adsorbed elements are deposited superficially on the periosteum, endosteum, and covering of the trabeculae (*27*, *28*) while Sr is deposited throughout the mineral portions in the bone salt, as shown by the radioautographs in Fig. 2.

The relations of the distribution coefficient, K_d, to the deposition in bone of radioelements are summarized in Table 3 on the next page.

Rate of Incorporation in Bone

Much indirect evidence indicates that the rate of passage of substances through the capillary endothelium *in vivo* is directly related to the diffusion rate *in vitro* (*29*). If diffusion rates govern the uptake of radioelements by bone, then, in accordance with the simple theory of diffusion in solids, the amount taken up should be a linear function of the square root of time. This can be expressed in terms of the parabolic diffusion law, as:

$$y = k\sqrt{t} \qquad (8)$$

in which y = the per-cent uptake of the element, t = time, and k = a constant that includes $\sqrt{D}$, D being the diffusion coefficient.

From the chemical properties of cationic radioelements, we would expect that the less basic elements would form larger aggregates by undergoing a greater degree of polymerization. Hence, the diffusion rates should increase as the *p*H of precipitation (Table 1) decreases. In rats, a plateau value in bone is reached in 24 hours with tetravalent plutonium, 6 hours with trivalent yttrium, and 2 hours with divalent strontium or radium. When Eq. 8 is applied to Ra, Y, and Pu uptake by the skeleton, following intravenous administration, a linear relationship is obtained nearly to the

TABLE 3

Relation of the Distribution Coefficient, K_d, to the Deposition of Radioelements in Bone

Basic Nonhydrolyzable Radioelements		*Hydrolyzable Radioelements*	
Ion Exchange Mechanism		Colloidal Adsorption Mechanism	
Physical-chemical Behavior	*Corresponding Biological Behavior*	*Physical-chemical Behavior*	*Corresponding Biological Behavior*
Uptake by exchanger is function of effective mass of bone, K_d remains constant	Uptake by skeleton of young growing animals greater than for adults	Uptake by exchanger largely independent of effective mass, K_d varies nearly proportionately	Uptake by skeleton independent of age of animal
Uptake governed by mass action principle, i.e., K_d decreases as concentration of cations in solution phase increases	Uptake by skeleton affected by changes in calcium metabolism or induced changes in bone	Uptake independent of cations, i.e., K_d remains constant despite changes in solution cation concentration	Uptake of skeleton unaffected by calcium metabolism, or induced changes in bone
Uptake is reversible, involves all exchange sites	Rate of elimination of fixed radioelements significant in man's life span Shift from initial site of deposition in bone as shown by radioautographs Diffuse distribution in bone throughout the mineral structure	Uptake is irreversible, primarily surface adsorption	Rate of elimination of fixed radioelements is negligible No shift from initial site of bone deposits as shown by radioautographs Distributed on bone surfaces, mainly periosteum and endosteum and covering of the trabeculae

plateau value (*29*). See Fig. 3. Such agreement is actually better than could be expected from the simple theory.

Blood Disappearance

Qualitatively, the percentage of the tracer cation that leaves the circulation within the first 5 minutes appears to increase with a decreasing tendency of the radioelement to polymerize at the *p*H of blood. In other words, the rapidly disappearing fractions represent diffusible ions that had not yet polymerized. The rate of polymerization or aggregate formation increases with decreasing basicity.

Two types of particles leave the blood very rapidly, those that are very large (~5,000 A.U. dia.), and those that are small diffusible ions (<10 A.U. dia.). All other particles leave the blood at rates corresponding to their diameters—the smaller the diameter of the polymer or aggregate, the slower is the rate of disappearance. Figure 4 is a diagram of these observations.

Some important experimental data bearing on this problem are given in the report by Dobson *et al.* (*30*). We have assumed that particles less than 10 A.U. in diameter represent simple molecularly dispersed systems, while those that are greater than 5,000 A.U. are microscopically visible (p. 2 of ref. *31*). Therefore, it was assumed that these two extremes represent the rapidly disappearing fractions. It should be noted that those large non-ultrafilter-

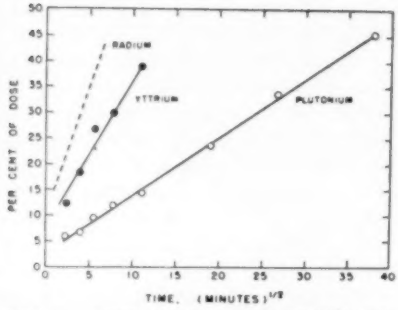

FIG. 3. Rate of uptake of Pu^{239} (IV), Y^{91}, and Ra^{226} by the skeleton of the rat according to the parabolic diffusion law. The points plotted represent the average value obtained from two rats at each time interval. The Ra^{226} data were taken from Norris and Kisieleski (*32*). After Schubert *et al.* (*29*)

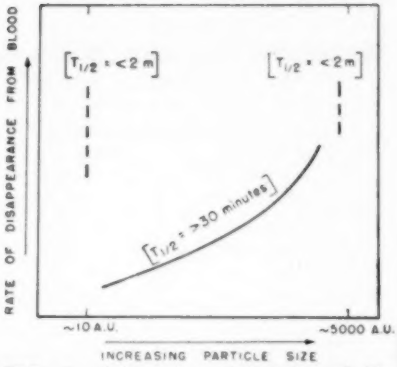

FIG. 4. Schematic relationship of the particle size of injected radioelements to the rate with which they disappear from the blood stream following intravenous injection

able particles which disappear from the blood rapidly seem to be cleared from the blood in a single passage through the liver (*30*).

Studies with Pu (IV) and Y (III) showed that by 5 minutes after intravenous injection 50% of the Pu and 80% of the Y had disappeared from the blood (*29*). Twelve hours later, 20% of the injected Pu was still being circulated, while the Y content had decreased to 0.7%. With alkaline earths such as Sr^{++}, Ra^{++}, and PuO_2^{++}, more than 90% of the injected dose disappears from the blood within 5 minutes (*32*). A semilog plot of data on Pu (IV) and Y is given in Fig. 5. The slow-leaving Pu fraction ($T_{1/2}$ = 8.4 hours) appears to represent particles with diameters of 40–50 A.U. (*29*).

Since the size of the aggregates formed increases with increasing concentration of the element, injection of larger amounts of the radioelement usually results in a less rapid disappearance.

Certain elements such as lead tend to concentrate in the blood cells rather than in the plasma fractions. As a result, the disappearance rates are more complex. Carrier-free Pb^{210} disappears

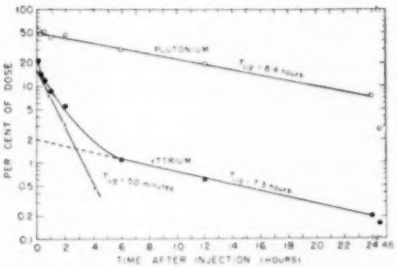

FIG. 5. Disappearance of Pu^{239} (IV) and Y^{91} from the circulating blood after intravenous injection of the soluble citrate salts at *p*H 6.5. Circles represent average values obtained from two rats at each time interval. Straight line drawn through the X points was obtained by subtracting extrapolated dotted line from curved portion of the yttrium curve. After Schubert *et al.* (*29*)

from the cells with a half-life of about 30 hours and from the plasma with a half-life of about 40 minutes (*33*). Interaction with plasma proteins is another factor which may modify disappearance rates.

Soft-Tissue Deposition

Colloidal substances are readily taken up by the reticulo-endothelial cells of tissues such as the bone marrow, spleen, and the Kupffer cells of the liver. It

TABLE 4*
Distribution of Zirconium Colloids in the Rabbit†

Organ	*% of injected dose Zr in organ*	
	$T_{1/2}$ = < 2 min	$T_{1/2}$ = 210 min
Bone + Marrow	9.3	43.0
Liver	85	23.0
Spleen	3.2	3.0
Muscle	1.3	17.0
Lungs	1.0	—
Kidney	0.3	14.4

* Data taken from Table 2 in ref. *30*.
† The colloid was zirconium-oxide suspended in sodium lactate (*62*). The two particle sizes are identified by the different half-times of disappearance from the blood.

is to be anticipated, therefore, that those radioelements with colloidal tendencies would be deposited to the greatest extent in the soft tissues. This is the case for those elements such as Zr, Po, Pu, rare earths, etc., when injected as an uncomplexed soluble salt. On the other hand, under the same conditions, only very small amounts of the alkaline earths are deposited in these tissues.

The relative distribution of colloidal substances depends on the particle size (*30*). The very large colloids, those that disappear very rapidly from the blood stream ($T_{1/2}$ = ~2 minutes) are deposited mainly in the liver and spleen. The colloids of smaller particle size are deposited primarily in the bone marrow and spleen, secondarily in the liver.

Once deposited, the excretion is very slow, and the organ distribution remains unchanged, and may remain so for years unless the compound is moderately soluble. Nearly four years after intravenous injection of Pu to a dog, the per-gram concentration of Pu in the liver was about one-half that of the skeleton (*33*). Dobson's data (*30*) on the distribution of zirconium colloids with different blood disappearance rates are given in Table 4.

Actually, the initial distribution pattern for any radioelement can be altered. For example, if strontium is injected as a simple soluble chloride salt, the distribution will be that of a typical alkaline earth, i.e., high skeletal and low soft-tissue deposition. However, if injected as an insoluble phosphate, Sr will localize in tissues such as the liver and spleen, the relative distribution, as mentioned above, being governed by particle size. Similarly, P^{32} can be directed into the liver and spleen (*34*).

When beryllium tracer is injected intraperitoneally as the chloride, about 16% is taken up by the pancreas and 5% by the liver (*35*). After intravenous injection, less than 0.1% is in the pancreas and more than 50% is in the liver.

It is of interest to compare the metabolism of Be^7 given as the chloride to that given as a citrate complex (Table 5). The higher skeletal and lower soft-tissue concentrations found after injection of the citrate salt are quite characteristic of amphoteric or hydrolyzable elements. Similar findings are reported in the case of Th (*23*) and gallium (*36*), for example.

In some cases, the fraction of an injected tracer deposited in the liver may differ even though the amount per kg body weight administered is the same. This could arise in the case where the concentration of tracer injected is different. Consequently, the particle-size distribution will fall in different ranges because the size of colloids produced is often dependent on the relative concentrations of the reacting solutions.

The uptake of ionic cations such as Sr by the liver reaches a peak and then decreases slowly. The same is true for colloidal particles, except that the rate of elimination from the liver is usually very slow, depending on the

innate solubility. If a colloidal substance is injected as a soluble complex ion, then its uptake by the liver will resemble that of an ionic cation. However, it leaves the liver largely as a colloid because the complex is destroyed. The colloids that are released into the blood stream are rapidly returned to the liver so that a second peak should arise. This, indeed, has been observed following the injection of gallium citrate (*37*).

Body Elimination of Radioelements

The two main routes of elimination of nonvolatile radioelements from the body are the urine and feces. In the case of an absorbed element, the ratio of the amount excreted in the urine to that in the feces is fairly constant, varying from about one to ten depending on the element and the time after exposure. However, following ingestion or inhalation of radioelements the principal route of excretion is usually via the feces.

The elimination of a radioelement is often stated in terms of the effective half-life, $T_{eff.}$ (*38*):

$$T_{eff.} = \frac{T_p \times T_b}{T_p + T_b} \qquad (9)$$

in which $T_p = 0.693/\lambda_p$, where λ_p = radioactive or physical decay constant, and $T_b = 0.693/\lambda_b$, where λ_b = biological rate of elimination constant (constant over a limited time interval only). When T_p is very large, then the effective half-life is equivalent to the biological half-life. No change in λ_p takes place, but λ_b changes with time. However, these changes are quite slow so that urinary excretion rate, while slowly decreasing over a period of months and years, appears to reach a steady plateau value (i.e., apparently constant λ_b) when examined during shorter time intervals.

Radioelements that are excreted via the kidney represent *absorbed* fractions. Those that appear in the feces may represent both absorbed and unabsorbed fractions; the latter fractions usually come from the gastro-intestinal tract as a result of oral intake or indirectly from inhalation. For purposes of estimating body content, it is very important to distinguish the two.

TABLE 5*
Comparison of Metabolism of Carrier-free Beryllium, Be^7, in Rats†

	Be as Chloride	*Be as Citrate*
Tissue	% inj. dose	% inj. dose
Carcass	35.7	35.1
Femur	0.90	1.82
Liver	16.0	0.76
Pancreas	5.4	< 0.1
Spleen	0.82	< 0.1
Kidneys	0.43	< 0.1
Urine	30	48
Feces	10	4

* After Schubert and White (*35*).
† Given intraperitoneally as citrate complex and as uncomplexed soluble forms. The animals were sacrificed 8 days after injection in the case of the Be-chloride rats and 12 days in the Be-citrate rats.

The absorption of insoluble compounds from the lung, for example, may be so slow that individuals who are found to have large amounts of a radioisotope in the feces may have little or none in the urine. Yet if the amount of isotope present in the lung were completely absorbed, the urine count would be quite marked. Since inhalation is a common route of exposure, it is well to examine the elimination of radioisotopes from the lung.

Metabolism after Inhalation

Following the inhalation of either smokes, aerosols, or dusts containing radioisotopes, the material appears in the lungs and the upper respiratory tract. The cilia of the naso-pharynx, the lower part of the vestibule of the larynx, the trachea, and especially the large bronchioles propel material toward the mouth where it is swallowed,

ultimately appearing in the feces (*39*). Besides the ciliary action, the peristaltic motion in the bronchioles, the cough reflex, and the nasal hair remove significant amounts of material (*40*). Certain cells (the dust or septal cells) are said to transport particles to the ciliated surfaces of the respiratory bronchioles, where they are subsequently removed by ciliary action. The review by Robertson (*41*) summarizes the ways in which foreign substances are eliminated from the lungs.

Material not removed mechanically can leave the lung in two ways. The foreign particles may be engulfed by phagocytes and carried into the lymphatic system of the lung by migration through the alveolar lining, or they may be dissolved and absorbed into the blood stream.

The degree to which the different particles reach various parts of the lungs depends on their particle size. In general, the depth of penetration into the lung increases with decreasing particle size while the fraction retained increases with increasing particle size (*42*). Those particles below 5 microns ($< 5 \times 10^{-4}$ cm) in diameter are exhaled to a considerable extent (*43*). (The nose will filter out almost all particles greater than 10 microns in diameter and about 95% of particles exceeding 5 microns.)

Several studies on the rate of elimination of radioisotopes from the lungs and their subsequent appearance in the feces, urine, and tissues have been made (*44*, *45*, *46*, *47*, *48*). The results of radioautograph and analytical studies are summarized (pp. 337–338 of ref. *46*) as follows:

> "Radioautographs of pulmonary tissue . . . revealed that, immediately after inhalation, the active material was deposited in both the bronchial tree and the air sacs. Within one day, the material in the bronchial tree vanished; presumably it was moved out by ciliary action and swallowed in the sputum. The large fraction remaining in the lungs was quite evenly distributed throughout the alveoli, and the pattern of distribution showed little change with time. The slow and continued release of active material from the alveoli probably also took place primarily by way of the bronchial tree. This deduction is based on the observation that the fecal excretion of plutonium and the fission product mixture closely corresponded to the elimination of these materials from the lungs."

The chemical properties of an inhaled radioelement affect markedly the rate of elimination and extent of absorption from the lung. The absorbed fraction of a very insoluble substance is slight, and hence the amounts retained by the lung are sources of intense local radiation. Relatively soluble substances may not be removed by mechanical action but will be dissolved and rapidly eliminated from the lung by passage into the blood stream.* Therefore, the amounts deposited in the skeleton and other tissues may be high.

In general, many of the elements listed in Table 1 with a precipitation *p*H of about 6 or less will be converted to relatively insoluble forms in the lung even when inhaled as a soluble salt. The relative amount transferred from the lung to the skeleton and other tissues is expected to be roughly proportional to the precipitation *p*H; the lower the precipitation *p*H, the less will be found in the tissues.

The efficiency of the lung in removing fission products is shown by Fig. 6. The rate of incorporation into the skeleton is also shown.

An illuminating sidelight illustrating the efficiency of the lungs for removing insoluble dusts in man is given by L.

* Inhalation of soluble compounds of radioelements such as Sr is nearly equivalent to an intravenous injection (*44*). The same situation holds for evaporated polonium inhaled by a rat (*49*).

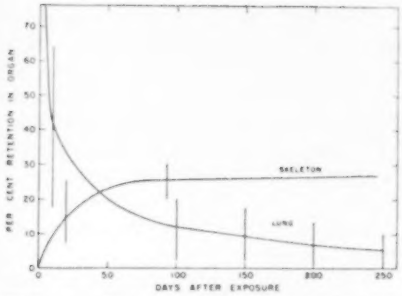

FIG. 6. Rate of elimination from rat lungs and rate of incorporation in the skeleton of inhaled aerosols containing hydrolyzable radioelements (Pu, Zr, Pa, Ce, Y, and mixed fission products) administered as smokes and sprays of soluble and insoluble forms (*44, 45, 63*)

Marinelli (private communication). He estimates that the total amount of dust inhaled throughout an anthracite coal miner's lifetime is 3,190 grams, consisting of 86% coal and 14% siliceous material. The average amount of dust found upon autopsy in the lungs of 27 anthracite miners was 25.5 grams or 0.8% of the estimated amount of inhaled dust [British Research Council Special Report 250, p. 69 (1945)].

Treatment of Radioelement Poisoning

The incorporation of long-lived radioelements by the body constitutes a potential danger as shown by the numerous cases of radium poisoning which occurred in the 1920's and early 1930's. Many attempts have been made to increase the rate of excretion of radioelements and to relocate deposits within the body to less radiosensitive areas. Little success follows treatment when the deposits have been fixed for several months.

One method for treating radioelement poisoning, particularly by Pu, is to induce formation of new, nonradioactive bone over Pu deposits so that the short range alpha radiations cannot reach the living cells (*27*). "This was accomplished by placing rats which had been injected with plutonium for one month on a regimen designed to produce bone resorption, and then changing them to an optimum diet calculated to restore the bones with new nonradioactive bone." Radioautographs revealed that a dense deposit of plutonium was buried deep in the shaft. This method has not been tested by toxicity studies in animals.

Van Middlesworth (*50*) attempted to mobilize Pu deposits in experimental animals in such ways as by induction of severe phosphate deficiency, vitamin C deficiency, protein-free diet, and injection of methionine and cysteine. No significant changes in either the distribution or excretion of Pu followed these treatments.

The ability of cations to form soluble complex ions has often been tried as a means of facilitating the excretion of radioelements through the membranes of the kidney and bowel. Citric acid, for example, has been used in the treatment of lead (*51, 52*) and uranium poisoning (*53, 54*).

A knowledge of the degree to which a complex-forming agent reacts with a cation to form a complex ion can be used to evaluate its possible usefulness for removing radioelements from the body.

Let us consider, for example, the effect of citric acid on radium excretion. From the measured association constant between Ra^{++} and citric acid at physiological *p*H and ionic strength, it has been calculated (*55*) that normally only 0.4% of circulating radium could be bound by citrate and hence available for excretion through the membranes of the kidney and bowel. In chronic radium poisoning in humans, the blood content averages 0.03% of the total amount fixed in the body. The kidneys filter about 180 liters of blood every 24 hours. Taking the average total blood volume to be 5.4 liters, we would expect that the amount of radium excreted via

the kidneys would be $(180 \times 0.03 \times 0.004)/5.4 = 0.004\%$ per day. This figure is about 10 times greater than the observed values (*56*). Hence it could be concluded that of all the radium passing through the kidney tubules about 90% is reabsorbed—which is a reasonable figure.

The effect of citrate administration as a means of increasing radium excretion can now be estimated. If 50 ml of a 3% sodium citrate solution is injected intravenously, the citrate-bound radium in the blood may be increased to about 2%. Even if such a level could be maintained* continuously for 24 hours (which is not the case), the urinary excretion of Ra could increase only five fold since the normal level of daily urinary excretion in long-standing cases is only about 0.0005% of the fixed body content (*56*).

The administration of citrates shortly after exposure to animals injected with radioisotopes of Th (*23*) and Be (*35*), for example, increases excretion and reduces soft-tissue concentration. However, the amount of the element deposited in the skeleton may be increased to a marked extent, as was observed with Be. The effect is even more marked when the radioisotope is injected initially as a citrate complex, as was found for Be (Table 5) and gallium (*36*).

It is apparent that the use of complex-forming agents for the treatment of radioelement poisoning may enhance the potential damage from radiation. This situation is in contrast to that involved with nonradioactive metal poisons such as lead where deposition in the skeleton reduces or eliminates the symptoms of metal poisoning.

However, complex-forming agents do not affect excretion or distribution of a hydrolyzable element like Th or Be which has been fixed in the body. Toxic levels of anions such as citrate are usually needed to dissolve the hydroxides of hydrolyzable elements as compared with the smaller amounts needed to prevent the precipitation of the hydroxides.

The metabolism of elements such as plutonium, the other transuranics, and the rare earths can be affected by the use of large amounts of nontoxic, nonradioactive elements such as salts of zirconium, titanium, and other hydrolyzable elements (*58*, *59*).

For example, during the 24-hour period following an intravenous injection of a soluble salt of Pu, only 1% of the injected dose is excreted in the urine. The administration of zirconium citrate to rats one hour after the Pu injection increases the 24-hour excretion level of Pu to as much as 60% of the injected dose, and decreases the amount of Pu deposited in the skeleton from 70% to 15% (*59*).

Zr salts and those of other hydrolyzable elements remove Pu or Y and similar radioelements by adsorption of the circulating radioelements on the colloidal aggregates which are necessarily formed by the injected Zr in the blood stream. The subsequent disposition of the radioelement is then related to the distribution pattern of the carrier salt. This, in turn, depends on the latter's particle-size distribution. A discussion of these points is given elsewhere (*33*).

In short-term experiments using moderate doses of Zr, no radioelement is actually removed from the skeleton;† instead, deposition is prevented by

* An elevation in the plasma citrate level would be expected to cause an increased urinary excretion of calcium. This was shown in experiments of Gomori and Gulyas and of Chang and Freeman (*57*). Repeated injections of citrate into puppies and rats by Gomori and Gulyas also led to osseous changes because of decalcification (*57*).

† Appreciable removal of Pu already deposited in the skeleton, spleen, and liver of rats has been obtained (*33*). The action is related to the fact that the rate of passage of the deposited radioelement from tissue is enhanced when blood level is maintained below normal.

removal of the circulating radioelement. Hence, it is possible to "freeze" the concentration of radioelements at whatever level it happens to be at the time treatment began.

In experiments with the effect of Zr on Pu and Y, the following observations were made (*59*): The amount of Pu or Y excreted in the urine was proportional to the size of the dose of Zr. The reduction of skeletal content of Pu was nearly independent of the amount of Zr injected, but that of Y was increased with increasing dosages of Zr. Liver retention of Pu was increased and then decreased with increasing dosages of Zr, but the Y content of the liver was little affected. Three days after injection the blood level of Pu in the Zr-treated rats was much lower than the control level. The amount of radioelement excreted in excess of that removed from the blood was derived from both the skeleton and the other tissues of the carcass.

The efficiency of Zr salts in minimizing Pu deposition is related to the chemical state of the Zr, as might be expected if the mechanism of Pu removal is by an adsorption mechanism. It has been shown that colloidal solutions of Zr citrate are much more effective in minimizing bone deposition of Pu than are completely ionic solutions. Furthermore, because residual amounts of Zr circulate in the blood for several days, it has been possible to minimize deposition of Pu in the skeleton by injection of Zr several days before Pu exposure. The toxicity of zirconium salts is exceedingly low (*60*).

The conclusion to be reached is that, in case of entrance of radioelements into the body, treatment should be started as soon as possible. In the case of excessive amounts of orally ingested radioelements, the appropriate treatment should minimize absorption from the intestinal tract (by use of precipitating agents) and hasten elimination. Fortunately, the adsorption of hydrolyzable radioelements such as Pu, Zr, and the rare earths from the gut is exceedingly small (<0.01%).

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"We need to maintain scientific perspective, even in times of emergency. Naturally, we need to put science and technology to work to solve immediately pressing problems. We need to develop materials, processes, machines and instruments to meet urgent needs. But we need more; we need basic research also.

"There should be no moratorium on basic research, even in times of stress, for research is the living source of progress. Research must provide a reservoir of facts and principles on which procedures and processes can be based when necessity arises."

—E. C. Stakman, "Science and Human Affairs," presidential address, AAAS, Cleveland, Ohio, Dec. 28, 1950

High-Energy Neutron Threshold Detectors*

Energy-dependent neutron effects are often more easily studied if threshold detectors are used to define the energy range of detection. Practical application data on choice of detector, analysis procedures, and specific uses are compiled

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MEASUREMENTS involving detection of neutrons are frequently complicated by the fact that, whereas the effects being measured are strongly energy dependent, most neutron sources yield a continuous energy distribution. In such cases, it is desirable to measure the flux, not of all neutrons as given by conventional neutron counters, but only of neutrons within a certain energy range.

Perhaps the simplest method of doing this is by exposing to the beam a substance which, when bombarded by neutrons above a certain energy (threshold), undergoes nuclear reactions whose end products are radioactive. After the bombardment, the activity is measured by standard counting techniques to give an indication of the flux of neutrons above the threshold. Such a substance is known as a "threshold detector."

Choice of Detector

The following factors must be considered in choosing a substance to be used as a high-energy neutron threshold detector:

Threshold. In the energy range from 1 to 25 Mev, and with neutron fluxes achieved to date, threshold detectors must depend on $(n,2n)$ or (n,p) reactions. Thresholds of the first type are obtained from mass measurements or by multiplying the measured (γ,n) threshold (*2*) by the factor $(A + 1)/A$ to correct for center of mass (A = atomic weight); these thresholds range from 9 to 21 Mev.

Energetic thresholds of (n,p) reactions can be calculated from the maximum beta energies of the ensuing beta decay and are found to lie generally between 0 and 3 Mev. However, the Coulomb barrier that must be penetrated by the emitted proton causes the cross section for (n,p) reactions to be very small near the energetic threshold. With the exception of phosphorus and sulfur, which will be discussed later, threshold detectors depending on (n,p) reactions are therefore of limited value for quantitative purposes.

Half-life. The beta-decay half-life of the product nucleus must be such that the counting can begin within about two to four half-lives after bombardment (depending on the intensity) and be continued for at least two half-lives. For example, if storage place is limited, it would be undesirable to use a detector with a 14-day half-life, which must be kept for about a month. On the other hand, if there is considerable equipment manipulation at the end of the bombardment, or if there are other

* Theoretical and experimental bases for the statements in this article are presented in reference *1*.

short-lived activities to be counted, a long-lived detector might be convenient.

In general it is not good practice to use a detector with a half-life much shorter than the bombardment time, as only the neutron flux at the end of the bombardment is measured. If a detector with a half-life many times longer than the bombarding time is used, there may be insufficient activity induced for practical work, although detectors with a half-life 500 times the bombarding time have been successfully used.

Other activities. Before choosing a substance for a threshold detector, one must examine the activities of the products of (n,γ), $(n,2n)$, and (n,p) reactions* in all isotopes of all elements in the substance to be certain that none of these have half-lives comparable with (roughly, within a factor of five) that of the interesting activity. For example, the 10-minute activity in copper due to $Cu^{63}(n,2n)Cu^{62}$ is difficult to use because of the 5-minute activity in Cu^{65} from $Cu^{65}(n,\gamma)Cu^{66}$. Similarly, great care must be exercised in selecting the chemical compound to be used. Organic compounds, if sufficiently stable, are suitable because the activities induced in carbon and oxygen are weak for most neutron sources. Calcium, on the other hand, is often associated with impurities which give considerable trouble over a wide range of (short) half-lives.

Physical, chemical properties. Substances which take on water from the air are often unsuitable because the absorption of beta rays changes during the counting, causing difficulty in identifying the decay curves and in correcting for the absorption. In addition, quantitative results are difficult to obtain, for they involve a knowledge of the number of atoms of the interesting elements, and this is obtained by weighing the dry substance.

Substances which vaporize at room temperature (e.g., iodine) must never be used, as the radioactive vapors permeate the counting chamber, making the measurement meaningless and giving a high background for future uses of the counter.

Due consideration must also be given to the chemical properties of the substance to be used. For example, dried acids will absorb enough moisture from the air to attack metal counter pans.

Tables 1 and 2 list some convenient threshold detectors and the necessary information for their use.

Procedures

The quantity that is measured by a threshold detector of threshold B is

$$I = \int_B^\infty F(E)\,\sigma(E)dE \qquad (1)$$

where $F(E)$ is the flux of neutrons with energy E and $\sigma(E)$ is the cross section for the reaction. From the definition of cross sections and the radioactive decay law, I is given by

$$I = \frac{\lambda n_0}{Q[1 - \exp(-\lambda\tau)]} \qquad (2)$$

where λ = the decay constant of the activity ($= 0.693/\text{half-life}$), τ = the bombarding time, n_0 = number of radioactive atoms at end of bombardment, and Q = number of atoms of interesting isotope per unit area perpendicular to the beam. From elementary considerations

$$Q = 6.03 \times 10^{22} \times \frac{maA}{MS} \qquad (3)$$

where m = mass of the substance, a = atoms of interesting element per molecule of the compound, A = abundance of the interesting isotope, M = molecular weight of the compound, and S = area perpendicular to the beam. It is easily shown from the radioactive decay law that, at a time t after the bombardment

* If the detector is placed at a position free from thermal neutrons—an unusual situation—(n,γ) reactions will give little trouble. Also, if the maximum energy of the neutron is below ~9 Mev, $(n,2n)$ reactions will be impossible.

TABLE 1
Convenient $(n, 2n)$ Threshold Detectors

Original Nucleus	*Threshold (Mev)*	*Half-life*	*Convenient Material*	*K-Capture Correction**	*Approximate α (mg/cm²)†*	*Other Activities*
C^{12}	20.2	20.5m	Graphite	1.00	70	None
N^{14}	10.6	10.1m	Urea	1.00	100	20.5m carbon $(n,2n)$
O^{16}	16.5	2.1m	Cellophane	1.00	170	20.5m carbon $(n,2n)$
F^{19}	10.4	112m	LiF	1.00	40	None
P^{31}	12.3	2.55m	$(NH_4)H_2PO_4$	1.00	400	170m (n,p)
Cr^{50}	13.4	42m	Cr_2O_3	1.00	240	3.9m (n,p)
Ni^{58}	11.7	36h	Ni metal	3.0	40	2.6h (n,γ); 72d (n,p)
As^{75}	10.3	16d	As_2O_3	1.3	85	26.8h (n,γ)
Ag^{107}‡	9.6	24.5m	Ag metal	2.3	220	2.3m (n,γ); 13h (n,p)
Sb^{121}	9.25	16m	Sb_2O_3	3.0	160	2.8d (n,γ)
I^{127}	9.45	13.0d	NH_4I	7.0	75	25m (n,γ)
Pr^{141}	9.4	3.5m	Pr_6O_{11}	1.6	360	Long

* Calculated from $(F_\beta - F_k)/F_\beta$ where F_β and F_k are probabilities of β^+ emission and K-capture respectively.
† This quantity, defined in text, is used in correcting for self absorption.
‡ Especially convenient for most purposes.

$$\log\left(-\frac{dn}{dt}\right) = \log \lambda n_0 - \lambda t \qquad (4)$$

where $(-dn/dt)$ is the rate of decrease of the number of radioactive atoms, which is proportional to the counting rate, dN/dt.

Corrections. The following factors enter into the proportionality constant:

1. Correction for geometry is determined by the average solid angle subtended at the counter window by the detector. This can be obtained by a numerical integration, although it is usually supplied by the manufacturers of commercial counting apparatus.

2. Correction for backscattering of the betas from the pan containing the material. This must be obtained empirically, but is small except for high-energy betas and thin detectors ($\sim 10\%$ for $E_\beta = 3$ Mev with zero-thickness detectors).

3. Correction for self absorption of betas in the detector itself. This is almost always important since comparatively thick detectors (> 5 mg/cm²) are usually necessary. The present author achieved good success with the following correction method:

It has been found experimentally that betas are absorbed according to

$$f = \exp(-d/\alpha) \qquad (5)$$

where f is the fraction of betas getting through a thickness d, and α is a constant independent of the material and very closely correlated with the maximum beta energy of the activity. It can be determined experimentally by inserting various thicknesses of aluminum between the active substance and the counter window, or the approximate values in Tables 1 and 2 can be used.

Integrating Eq. 5 through the thickness of the detector gives:

$$\bar{f} = \frac{\alpha}{d_0}\left[1 - \exp\left(\frac{-d_0}{\alpha}\right)\right] \qquad (6)$$

where d_0 is now the total thickness of the detector.

4. Correction for absorption of the betas in the counter window (its thick-

TABLE 2
Convenient (n, p) Threshold Detectors

Original Nucleus	Energetic Threshold (Mev)	Effective Threshold (Mev)	Half-life	Convenient Material	Approximate α (mg/cm^2)	Other Activities
Mg^{24}	2.1		14.8h	Mg metal	250	10.2m (n,γ)
Al^{27}	2.1		10.2m	Al metal	160	2.4m (n,γ)
P^{31}	1.1	~3.0	170m	$(NH_4)H_2PO_4$	120	2.5m $(n,2n)$
S^{32}	1.0	~3.0	14.3d	Flower of sulfur	160	None
Ti^{49}	1.1		57m	Ti metal powder	190	3.0h $(n,2n)$; 44h (n,p)
Cr^{52}	2.8		3.9m	Cr_2O_3	350	42.0m $(n,2n)$
Fe^{56}	2.1		2.59h	Fe metal	250	None

ness is usually given by the manufacturer) and air can be calculated from Eq. 5.

Use of these corrections allows one to determine $(-dn/dt)$ from the observed counting rate dN/dt. A plot of the former versus time on a semilog scale gives, according to Eq. 4, a straight line of slope such that $(-dn/dt)$ decreases to half its value in a time equal to the half-life. Extrapolation of this line back to $t = 0$ (the end of bombardment) gives λn_0, the quantity needed to obtain I from Eq. 2. Suitable combination of Eqs. 2 through 6, leaves the bombarding time, τ, the mass, m, the area S, and the decay curve intercept at $t = 0$ to be obtained experimentally. [For thick detectors $(d >> \alpha)$, m is not important.]

When decay curves are plotted, longer-lived activities must first be identified and subtracted from the counting rate. This usually necessitates measurement of the latter over a period of several half-lives. Sometimes one or two readings at a very much later time, about 20–50 half-lives after bombardment, are necessary. Where shorter-lived activities are present, it is useless to do counting until these have decayed away. It is very important that a decay curve of the proper slope be obtained before any use is made of the data.

If Eq. 1 is to yield information about the absolute neutron flux, it is necessary to know the cross section as a function of energy. For $(n,2n)$ reactions in medium and heavy elements (atomic weight > 50), this is (3)

$$\sigma(E) = \pi r^2 \left[1 - \left(1 + \frac{E - B}{T} \right) \exp\left(- \frac{E - B}{T} \right) \right] \quad (7)$$

where r is the nuclear radius, B is the threshold, and T is the nuclear temperature at the energy of the incident neutron, E_n. Improved values for T will eventually be available (4), but Weisskopf's latest estimates (5) are represented by $T = (E_n/a)^{1/2}$ where a varies from 1.3 Mev^{-1} for an atomic weight of 55 to 6.5 Mev^{-1} for an atomic weight of 115.

It must be noted that the products of $(n,2n)$ reactions decay not only by beta emission, but also by K-capture. The theoretical corrections for this are listed in Table 1.

For $(n,2n)$ reactions in light elements and most (n,p) reactions, present knowl-

edge of cross sections as a function of energy is scanty. However, the (n,p) cross sections for S^{32} and P^{31} are available (*6*); because of a fortunate occurrence of resonances, these rise very steeply at about 3 Mev and are reasonably flat thereafter—a most ideal situation for threshold detectors.

Applications

Neutron fluxes. Given a knowledge of the energy spectrum, one can use any heavy $(n,2n)$ detector to measure the absolute flux by applying Eqs. 1 and 7 with the experimental value of I. However, a much better method for most purposes is to use sulfur or phosphorus, in which case the total flux above about 3 Mev can be found without a knowledge of the energy spectrum. This experiment consists of determining I experimentally and applying Eq. 1 with $\sigma = 0.32$ barns for sulfur (*6*) or $\sigma = 0.14$ barns for phosphorous (*1, 6, 7*). This method was used by Allen *et al.* (*7*), and was found to agree well with the results of other types of measurements.

Energy spectrum. Although there are better methods for determining neutron energy spectra (e.g., by use of photographic emulsions), a first estimate can be obtained by applying Eq. 1 with several different detectors. If the neutrons are obtained by bombarding any but the lightest elements, the most practical method is to assume a Maxwellian spectrum* and determine effective temperatures.

If the neutrons are obtained by bombarding a fairly light element with high-energy alphas or protons, a convenient method (*8*) is based on the facts that the angular distribution of neutrons is symmetrical about 90 degrees in the center of the mass system, and that the center of mass correction is very sensitive to the energy spectrum. The method consists of finding the relative activities, R, induced in identical threshold detectors placed at 0 and 180 degrees with respect to the incident charged-particle beam. A correction for center-of-mass solid angle is made (*9*) by employing the relation

$$R_c = R\left(\frac{\gamma - 1}{\gamma + 1}\right)^2$$

where $\gamma = (M_A/\sqrt{M_a}) \cdot \sqrt{E_n/E_a}$ and M_a = mass of incident particle, E_a = its energy, M_A = mass of target nucleus, and E_n = energy of neutron. Also, the effective temperature T of the spectrum—assumed to be Maxwellian—is determined by using the equation

$$R_c = \frac{M + T}{N + T} \exp\left(\frac{N - M}{T}\right)$$

where N and M are

$$\frac{N^{(+)}}{M^{(-)}} = \frac{1}{2}\left[\sqrt{2B} \pm \frac{\sqrt{2E_aM_a}}{M_a + M_A}\right]^2 \quad (9)$$

and E_a and M_a are the energy (in Mev) and mass (in atomic units) of the incoming charged particle, M_A is the mass of the struck nucleus, and B is the effective detector threshold. For $(n,2n)$ detectors, B should be about 1 to 2 Mev above the energetic threshold.

Angular distributions. Probably the best way to measure angular distributions of high-energy neutrons from charged-particle reactions is to use threshold detectors. This is true primarily because these detectors are very little activated by scattered neutrons.† However, corrections must be made for:

1. Neutrons scattered out of the beam by material between the target and detectors. This correction can be made by using the cross section for neutron scattering as πr^2 for material near the target or the detector, and $2\pi r^2$ for material far from each. (Elas-

(*Continued on page 45*)

* By Maxwellian spectrum is meant $F(E) = CE \exp(-E/T)$, where T is the effective temperature.

† High-energy elastically scattered neutrons are concentrated in the forward direction, and inelastically scattered neutrons will mostly have insufficient energy. For a detailed discussion see reference *11*.

Chemical Dosimetry of Ionizing Radiations

Chemical techniques were among the first used to measure radiation dose. Lately, after being long overshadowed by ionization techniques, they have found new importance. Local, integral and auto-integral dose can be measured accurately with liquids and gels by employing the methods described

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WHEN X-RAYS AND RADIUM came into medical usage, a need was created for quantitative determination of absorption of radiation energy in tissue-like systems.* After trial of many methods, ionization techniques were gradually accepted as the most practical way of meeting this need. With the advent of more intense radiation sources, however, chemical techniques have been found useful, especially since they obviate many of the difficulties associated with ionization dosimetry.

The name of "dosimetry" for this field of radiation physics stems from the field's medical origins. The amount of degraded radiation energy is known today as the "dose."

Soon after the need for quantitative measurement was recognized, various methods of dosimetry were proposed. Most were purely empirical in character; the distinction between total energy flux (intensity) and the rate at which that energy is absorbed (dose) was not always appreciated. For instance, neither the photographic plate nor the fluorescent screen absorbs all the energy from an X-ray beam of moderate wavelength, and therefore the blackening or brightness, as the case may be, is not a measure of the intensity of the beam. On the other hand, both will absorb more X-rays than a corresponding piece of biological tissue (the ratio depending on the wavelength) and hence are equally unsuitable for the measurement of dose. Various early chemical or quasi-chemical methods of dosimetry—production of F-centers (Holzknecht, 1902), liberation of iodine from iodoform solution (Freund, 1904), precipitation of calomel from a mixture of mercuric chloride and ammonium oxalate solutions (Schwarz, 1907), and barium platinocyanide pastille (Sabouraud-Noiré)—all suffered from similar disadvantages (*1*).

Ionization Dosimetry

Fundamentally, the ionization method has a direct bearing only on energy absorption in gases, but it was gradually accepted as the most convenient and practical method of dosimetry. The corresponding unit of dose appropriate to this method (the roentgen) was defined in 1929 and has remained substantially unchanged. In absolute units, when 1 gm of air is irradiated to a dose of 1 *r*, the energy absorbed is about 84 ergs.

The science of dosimetry has become

* These are extended systems, mainly aqueous, and have about the same effective atomic number as air, bone being an outstanding exception.

an essential part of radiation physics, though its medical terminology and relevance remains. Now that other methods of arriving at the dose are coming into use, it is convenient, especially for medical purposes, to refer them to the ionization scale using the figure 84 ergs/gm as the equivalent roentgen (rep).

Because of the finite range of the secondary electrons produced by X-rays in air, the energy actually absorbed from an X-ray beam at a point produces ionization in a large surrounding volume, and the dose in roentgens at the point can only be measured by collecting all these ions. For this reason dose determination by the direct measurement of ionization in air, which is implied in the definition of the roentgen, can only be realized experimentally under very limited radiation conditions, such as exist in the "free-air" chamber (*2*). The "air-walled" cavity chamber, in which the surrounding air volume is in effect compressed into a wall of similar atomic composition immediately surrounding the reference point, overcomes this difficulty.

The cavity-chamber method was first placed on a sound theoretical basis by Gray (*3*, *4*) following a similar suggestion by Bragg (*5*). Gray used the ionization method to determine the total gamma-ray energy emitted by Ra (B and C) and Th (B and C) and obtained fairly good agreement with calorimetric values (*6*). His arguments and his successful applications of the theory showed that the ionization method could be applied, within certain limits, to absolute measurement.

Gray showed that the ionization in an air cavity in an irradiated dense medium is proportional to the energy absorption per cm³ in that part of the medium directly surrounding the cavity. In precise terms

$$E_r = J_r W \rho$$

where E_r = energy absorbed per cm³ in dense medium, J_r = number of ionizations per cm³ in gas cavity, W = average energy required to form an ion-pair, and ρ = stopping power of dense medium relative to gas.

Practical "thimble" or capacitance chambers are designed to make use of this relation, although in practice departures of up to 20% from the theoretical relation are to be expected from an actual experimental chamber. This can be partly overcome by calibrating each chamber empirically over a range of wavelengths by comparison with the "free-air" chamber.

Although the ionization method of dosimetry has several practical advantages, especially the ease and accuracy with which the ionization current can be measured, it has recently come in for critical re-examination (*7*). Actually, the constants W and ρ in Gray's fundamental relation are somewhat dependent on the energy of the ionizing particle. In addition to errors thus caused, there are several experimental complications. The atomic number of the chamber material is of critical importance but cannot be adjusted to the correct value except by individual trial and error. The effect of chamber geometry and an unpredictable "wavelength effect" also involve appreciable sources of error. In addition, the measurement of integral dose (i.e. the total energy absorbed in a medium) can only be carried out by the integration of a large number of observations.

Other Measurement Methods

For these reasons, plus the fact that increasingly intense radiation sources are coming into use, more attention has recently been paid to other means of measuring radiation, using effects in solid or liquid media rather than in gases. Examples are the fluorescence method, used in the form of the scintillation counter for low-intensity radiation (*8*, *9*), and the luminescence method

TABLE 1

Variation of W with Energy of Ionizing Electron

Electron energy, kv	0.5	0.8	1.0	2	5	10	20	50	100	1,000	∞
W, ev/ion-pair	39.2	37.6	36.9	35.4	34.0	33.3	32.8	32.4	32.1	31.8	31.6
W, relative	1.24	1.19	1.17	1.12	1.07	1.05	1.04	1.024	1.017	1.005	1.000

for measurement of high-intensity radiation (*10*). Also in this category are various methods which consist essentially of the observation of the chemical changes produced by radiation using various methods of analysis, e.g., titration (*11*), colorimetry (*12*), conductivity (*13*), and *p*H changes (*14*). The production of color centers in crystals (*15*), and the blackening of the photographic emulsion (*16*)—both early methods of radiation measurement—have received fresh consideration.

Wherever the ultimate aim is the determination of the dose in a biological tissue, the ideal method of dosimetry should involve a system as tissue-like as possible. Failla has made an approach to the direct measurement of ionization in tissue using the "extrapolation chamber" (*17*), while Spiers has studied the absorption of X-rays in various tissues (*18*). It seems possible that a true "water-walled" chamber can now be constructed using gels to form the walls (*19*), but such a technique suffers from the fundamental difficulties of any ionization chamber.

From an early time it has been realized that if a reliable chemical method could be evolved which would give the absorbed energy in terms of the induced chemical reaction in a tissue-like aqueous system, such a method would be in many ways preferable to the ionization method. In particular, such a system is equally suitable for the determination of integral or local dose, irrespective of the shape or size of the irradiated body.

It appears that chemical changes are largely due to the *ionizations* in the aqueous systems; the contribution of the excited molecules, which are responsible for about half the total energy absorption, are unclarified at the present time. The chemical effect is proportional to the number of atoms ionized rather than the total energy absorbed. However, the energy required to produce an ion-pair is only slightly dependent on the energy of the ionizing particle for energies above a few kv (*20*) (Table 1). Such experiments as have been made on ionization in liquids do not suggest gross qualitative departures from values obtained in their gaseous state. We are probably justified in making the provisional assumption that W for water is almost independent of the energy. Thus, the factor of proportionality relating chemical effect to energy absorption is constant within 5 % for all electron energies greater than 2.8 kv and within 1 % for energies above 70 kv. Within these limits of accuracy, a suitable chemical method would provide a true measure of energy absorption.

In addition, from the biological angle, it seems very likely that the chemical changes produced by radiation will be a measure of the extent of biological action. Therefore, the biologically significant factor is the one determined by a suitable chemical method and not necessarily by total absorption.

Several of the chemical systems proposed for dosimetric purposes must be criticized because they include atoms of high atomic number and hence are only suitable for photons of high energy. It must be emphasized that for general utility a dosimetric system must have a mean atomic number [$\bar{Z} = \sqrt[2.94]{\Sigma nZ^{2.94}/\Sigma nZ}$] within at most 1 % of the value for tissue (soft tissue,

$\bar{Z} = 7.4$) or for water ($\bar{Z} = 7.42$) (*18*). The use of higher atomic number media (inorganic crystals such as potassium iodide, and liquids like chloroform) increases the chemical yield from a given flux of X-rays, but only by sacrificing the criteria for true dosage measurement.

The photographic method, which may be regarded as a special form of chemical dosimetry, suffers from the fact that the blackening is dependent on the specific ionization along the electron tracks. Its high sensitivity, however, means that it may be of value in protection work. It may also have uses in "mega-voltage" dosimetry, since the specific ionization of high-energy electrons is almost independent of the energy.

Chemical Techniques

It seems worthwhile to re-examine the scope and limitations of the chemical method. The chemical changes produced by ionizing radiations must be amenable to measurement with an accuracy at least as good as a few per cent, using techniques simple enough for ordinary laboratory routine. This sets a limitation on the smallest amount of chemical change which must be produced, and hence on the lowest measurable dose. In practice it is found that, since about 20–35 ev is expended in producing a primary chemical change in one molecule, the smallest dose which can be measured with the desired accuracy without utilizing chain reactions is of the order of a few hundred roentgens. For this reason chain reactions, giving a multiplication factor over the initial chemical change and thus increased sensitivity, have been the object of study [hydrogen peroxide (*21*), Eder's solution (*1*), chloral hydrate (*13*), etc.]. However, owing to the labile nature of the systems, the multiplication factor tends to be a function of the experimental conditions, and the increased sensitivity is counterbalanced by decreased stability and reproducibility.

In the absence of a really suitable chain reaction, which would be, of course, the best realization of the idea behind chemical dosimetry, several systems have been worked out which, while not sensitive to doses of less than a few hundred roentgens, give satisfactory results above this limit. Such a system must be stable in storage (i.e. not give a "blank" in the absence of radiation) and must keep its final reading after irradiation without further change.

The use of ferrous sulphate solutions was first suggested by Fricke and Morse (*22*) and developed to a refined technique by Miller (*11*, *23*). This method answers most of the preceding requirements and can be used to the lowest limit of chemical dosimetry. An alternative method using sodium benzoate or benzene as substrate has been suggested (*12*). This has almost the same sensitivity as the ferrous sulphate method, but has the additional advantage that both the initial solution and the products of irradiation can be kept for long periods without serious deterioration. Thus, for instance, it was possible to follow the chemical effect of the β-rays of P^{32} for 30 days.

Radiation action in aqueous solutions. All chemical methods of dosimetry utilizing aqueous solutions derive from the primary changes in water itself. Although the final result is a chemical change in the solute, we are thus dealing, in dilute solutions, with the absorption of radiation in water itself, thereby achieving the fundamental aim of dosimetry. It is generally accepted, in agreement with the theory of Weiss (*24*, *25*), that the primary ionization

$$H_2O \rightsquigarrow H_2O^+ + e$$

is followed by secondary processes.

From the H_2O^+, free OH radicals are formed

$$H_2O^+ \rightarrow H_{aq}^+ + OH$$

This process is exothermic owing to the hydration of the proton formed. The ejected electron may cause further

ionizations. When finally slowed down sufficiently, it may be captured by the water and lead to formation of H atoms

$$H_2O + e \rightarrow H_2O^-$$

$$H_2O^- \rightarrow OH_{aq}^- + H$$

The subscript *aq* denotes that the molecules are part of the condensed water system. The subsequent chemical reactions are oxidations and reductions due to the OH radical and the H atoms thus formed. These bring about further changes in the small quantity of suitable substrate, thus indirectly transferring the action of the radiation (*26*, *27*, *28*).

Thus, in the case of $FeSO_4$, the free radicals oxidize Fe^{++} to Fe^{+++} (*23*); in the case of benzoic acid, they enter into reaction with the eventual formation of hydroxy-benzoic acids (*29*, *30*). In these instances the chemical effects are due to the OH radicals. In the case of Fe^{++}, an electron transfer process occurs

$$Fe^{++} + OH \rightarrow Fe^{+++} + OH^-$$

while, in the case of benzoic acid, a dehydrogenation occurs

$$C_6H_5COOH + OH \rightarrow C_6H_4COOH + H_2O$$

which is followed by the formation of the phenolic body

$$C_6H_4COOH + OH \rightarrow C_6H_4(OH)COOH$$

The H atoms formed from the same primary ionization as the OH radicals may react with oxygen if present in the solution and give rise to further oxidation instead of reduction (*29*).

The chemical yield in these reactions is an accurately reproducible quantity and suitable for dose measurement. Important advantages of this technique are that yields are not critically dependent on *p*H, are only slightly dependent on concentration of reagents, and, as far as we have observed, are independent of temperature (Fig. 1).

Standardization. The most convenient way to standardize any chemical reaction is to measure the chemical change produced by a dose of radiation that has been accurately measured with

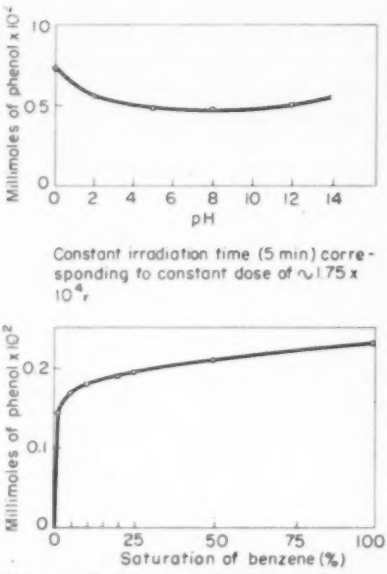


FIG. 1. Inflence of pH and concentration of benzene on yield of phenol

suitable ionization chambers, which must themselves be calibrated against an appropriate standard. An apparatus of the type shown in Fig. 2 was used for this purpose. The solution (usually ½ or 1 cm³) was contained in a very thin walled Perspex* tube which was held in a fixed position in the X-ray beam. Facilities were provided for filtering the X-rays, and various effective wavelengths could be achieved by a combination of filter and tube voltage adjustments.

The dose rate at the end of the irradiation tube was measured by introducing a very small ionization chamber of the type described by Farmer (*31*) and calibrated for the various radiation qualities against a substandard dosimeter which had itself been standardized by the National Physical Laboratory against the free-air chamber.

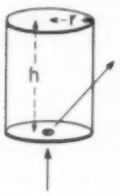
A monitoring ionization chamber coupled to an X-ray stabilizing system

* Perspex is the trade name for a transparent polymerized methylacrylate plastic.

(*32*) was of considerable assistance in achieving an accurately known dose.

Even in such a small volume of liquid it is necessary to apply corrections for the absorption and scattering of the primary radiation. In view of the experimental difficulties consequent on the small size, coupled with the fact that small errors in such corrections could not introduce any appreciable error into the final result, it was decided to calculate these correction factors. The mean primary dose was fairly easily calculated, being simply a matter of inverse-square attenuation plus exponential absorption. The scatter dose S, i.e., the amount of energy scattered by one element of the volume and absorbed in another, was approximated by a three-step calculation based on the relationships depicted below.

A. The energy scattered by $d\tau$ and absorbed in the cylinder shown at the right, where h and r are variable and $d\tau$ is an element of volume in the cylinder, was calculated, giving



$$S \cdot d\tau = f_1(h,r)$$

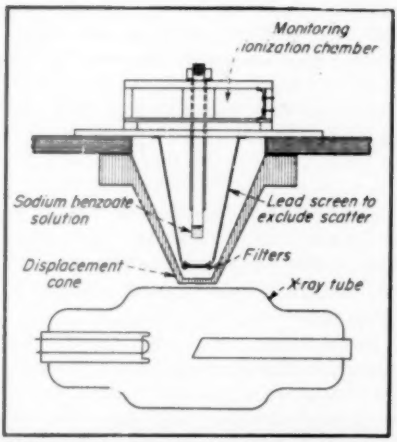
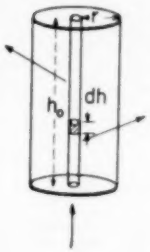


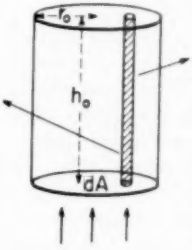
FIG. 2. Apparatus for standardization of sodium benzoate method of dosimetry

B. The result obtained in **A** was integrated to obtain the energy scattered from the filament dA (of which dh is an infinitesimal segment) and absorbed in the cylinder. Exponential irradiation of the filament was assumed, and, with r variable



$$S \cdot dA = f_2(r)$$

C. Knowing the way in which the scatter dose varied with the radius, it was possible to compute approximately "equivalent radii" for filaments off the axis of the cylinder and subsequently an "equivalent radius" for the whole cylinder. Substitution of this value in **B** yields



$$S = f_2(\bar{r})\pi r_0^2$$

In these calculations we followed Lamerton (*33*) in using the following approximation to the Klein-Nishina formula, which is a reasonably good fit and greatly simplifies the mathematics

Forward-scatter: $f(\theta) = A - B\cos^2\theta$

Back-scatter: $f(\theta) = A' - B'\cos^2\theta$

Chemical Procedure

To determine the dose, analytical-reagent sodium benzoate is dissolved in distilled water to make a 0.5% solution. The vessel to be irradiated is filled with this solution. After irradiation an aliquot of the solution is withdrawn. When the dose at a point is to be measured, 0.5 ml of irradiated solution is taken, and to it is added, in a test tube, exactly 0.5 ml of Folin's reagent (*34*), 1.5 ml 18% sodium carbonate solution and, according to the colorimeter cell used, a measured quantity of water (0.5–1.5 ml) to obtain enough solution

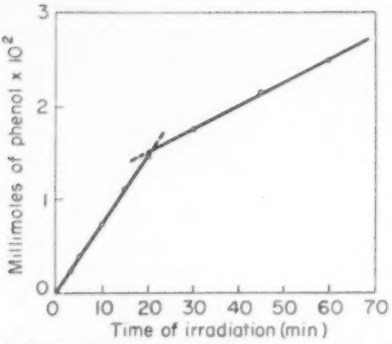


FIG. 3. Yield of phenol plotted against dose. Dose rate is 3,600 r/min. Note sharp break in curve when all oxygen initially present in the solution was exhausted

to fill the cell. The contents of the test tube are mixed by rotating and warmed for 15 minutes at 50° C on a waterbath, to obtain full color development. The test tube is then cooled under the tap and the liquid introduced into the colorimeter cell.

When integral doses in large volumes are measured, greater quantities may be withdrawn, and using the same procedure, greater extinctions* may be obtained, allowing use of smaller doses.

The color developed is due to the action of phenolic bodies on the reagent. Using this technique the depth of color developed is an accurately reproducible linear function of the dose up to about 60,000 roentgens (Fig. 3). As a purely arbitrary standard we chose salicylic acid, which gives the same color with the reagent, is quite stable, and can be obtained in great purity.

The observations were repeated over a range of radiation qualities and a similar experiment was performed with radium gamma-rays, using six 200-mgm radium tubes supported in a 2½-cm diameter circle. At the center of the circle was a Perspex tube (walls 2 mm thick) containing ½ cm³ of the irradiated liquid. The dose-rate was measured using the same ionization chamber as in the X-ray experiments.

Comparison of relative and absolute yields shows that within the experimental errors the chemical yield is a true measure of the energy absorbed. (Note that if the dose is expressed in roentgens there is a small "wavelength effect" because the energy absorbed in water per roentgen depends somewhat on the wavelength.) It was found (Table 2) that a salicylic acid concentration of 5.43×10^{-10} gm/cm³ was equivalent to a dose of 1 rep.

TABLE 2
Yields of Phenols on Irradiation

Relative Yields (0.5% sodium benzoate)

Wavelength, A.U.	0.015	0.12	0.18	0.23	0.32	0.41	0.46
Yield per rep	0.96	1.00	1.02	0.97	1.00	1.02	0.99
Yield per r	0.99	1.00	0.98	0.91	0.92	0.94	0.91

Absolute Yields

1 rep produces phenols equivalent to $5.43 \pm 0.15 \times 10^{-10}$ gm/cm³ salicylic acid (0.5% sodium benzoate)

or $5.21 \pm 0.20 \times 10^{-10}$ gm/cm³ salicylic acid (saturated benzene solution)

Applications

The test of any dosimetric scheme is in its practical application to the solution of specific problems, and the described systems have been applied in three ways:

Local dose. We have used the chemical technique to measure the surface backscatter which is obtained when an X-ray beam falls on a large block of water-like material. A shallow Perspex cell, diagrammed in Fig. 4, with a thin

* The term "extinction" is used in preference to "optical density" to which it is equivalent. It is defined as equal to $\log_{10} I_0/I$, where I_0 is the intensity of light in absence of absorbing substance and I is the intensity of light transmitted through the absorbing substance. The extinction is proportional to the concentration of the absorbing substance whenever Beer's law holds.

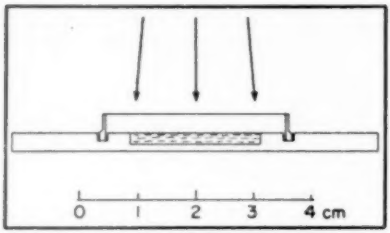


FIG. 4. Cell for back-scatter measurements

paper cover to exclude dust and prevent evaporation, was placed in the X-ray beam and the effect of bringing up a scattering phantom was studied. The results of this preliminary experiment are not of high accuracy, but are in agreement with ionization measurements in suggesting that the backscatter given by Mayneord and Lamerton is about 20% low (400-cm field; half-value layer, 1.2 mm Cu) (*35*).

Integral dose. The integral dose in the irradiation vessels used in our radiation chemistry work is now measured with the sodium benzoate technique as a routine procedure (*36*, *37*). With apparatus normally available in any chemistry laboratory, an over-all accuracy of about ±3% can be obtained. The dose distribution in one such vessel was also surveyed using an ionization chamber. Very satisfactory agreement with the chemical method was obtained, the two values differing by only 3%.

Auto-integral dose. The same technique was extended to the measurement of auto-integral dose, i.e., the integral dose given to a body by its own radioactivity. This experiment may be of interest in view of attempts to calculate the dose in bodies of irregular shape, such as are met with in biological experiments (*38*, *39*, *40*). The curves in Fig. 5 show the accumulation of phenol in aqueous benzene solutions containing P^{32}.

The experiment may also be regarded as a determination of the mean β-ray energy, on which the auto-integral dose

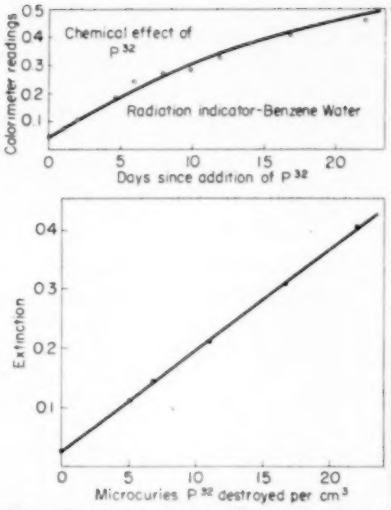


FIG. 5. (Top) Production of phenol through decay of P^{32} in solution. (Below) Relation of dose to μc P^{32} destroyed

depends, and we have carried out this experiment, under standard conditions, using P^{32}. The solution containing the radiophosphorus in the form H_3PO_4 was first freed of traces of reducing impurities by the addition of pure hydrogen peroxide (100 volume) and of bromine water, which were subsequently driven off by heating. The phosphoric acid containing the isotope was then introduced into an aqueous solution of benzene, which was then divided between several spherical flasks covering a range of sizes (10–100 cm^3). With extrapolation to infinite radius, allowance could then be made for the escape of β-particles from the solution. At intervals over 2 or 3 days, the yields of phenol were measured and thus the dose determined. The activity of the solution was determined by quantitative dilution and comparison with a calibration sample supplied by the Atomic Energy Research Establishment, Harwell, England, using a liquid counter (*41*). This standard solution was based on measurements made with a counter of defined

solid angle (*42*). Using values for the activity as thus determined, our results gave an average value for the mean β-ray energy as 0.66 Mev. This agrees fairly satisfactorily with the generally accepted value of 0.69 Mev. In view of difficulties in the absolute assay of radioisotopes, this approach would seem to be of special interest.

Chemical Dosimetry Using Gels

While these chemical methods utilizing solutions do answer some of the requirements of dosimetry in tissue-like systems, and in particular may be used for the measurement of dose at a point as well as integral dose, a further improvement can be obtained by the use of solid systems, which would give the three-dimensional distribution of the radiation in a field.

Some of the requirements of such an improved system are fulfilled by a method developed recently, in which use is made of the change in the color of dyes dissolved in gels (*43, 44*). These color changes can be observed directly, without the addition of any further reagents or developing. The gels are prepared in our experiments by adding to hot water sufficient gelatine or agar (1–10%) to make it solid on cooling, and sufficient dye, e.g. methylene blue (0.002%) to obtain a convenient depth of color. Accordingly, their composition and average atomic number are practically identical with those of tissue, and can be adjusted to any required value by suitable additions. On irradiation, color changes occur which can be utilized for dosimetry.

The primary processes involved in the chemical action of radiation in gels are identical with those in solutions, and owing to the fact that diffusion constants in these gels are very nearly the same as in water (*45*), the secondary chemical reactions will presumably be similar too, with the difference that effects of convection and mixing are eliminated. Therefore, the chemical products will be confined close to the point where they were formed for periods long enough to obtain three-dimensional pictures of the dose distribution.

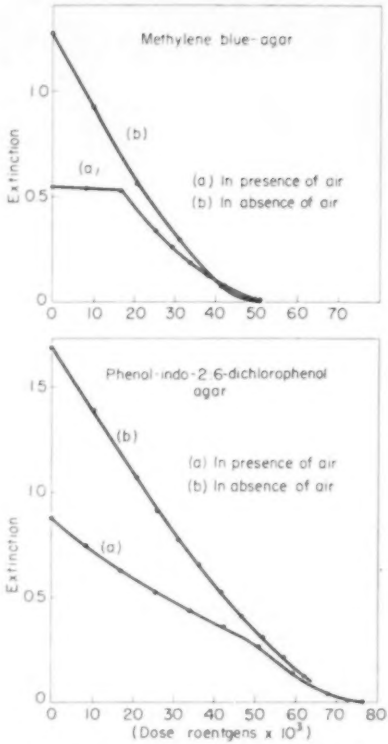


FIG. 6. Irradiation of indicator gels

From the point of view of dosimetry, these systems can be adjusted so that the change of extinction is a linear function of dose given. Suitable shapes can be formed by the use of appropriate Perspex cells to obtain either the three-dimensional distribution of radiation in a field, or the dose at any selected point.

In the systems investigated by us until now, use has been made of the reducing action of the ionizing radiations, rather than their oxidizing properties. The detailed mechanism of the action of radiations in these gel systems is not yet perfectly understood,

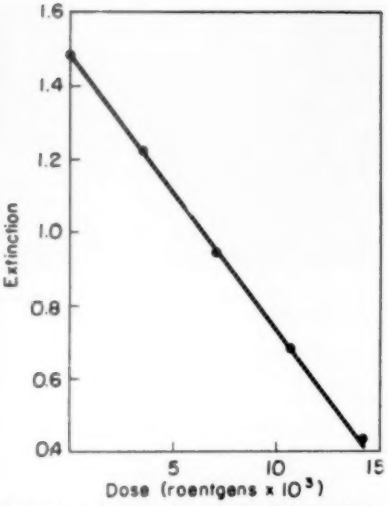


FIG. 7. Irradiation of methylene-blue-agar gel containing benzoate. Note increased sensitivity

but apparently, in the presence of a suitable acceptor like the dyes used, even a large concentration of other potential acceptors such as gelatine or agar does not materially decrease the yield in terms of molecules of dye reduced. It is only when other similar acceptor dyes, or other substances known to be good electron acceptors (e.g., oxygen) are present that competition occurs. This competition involves apparently reversible processes either for the electrons themselves, or for the hydrogen atoms formed from them.

Thus, in the presence of oxygen in the gel, the extent of the reduction of the dye depended on its oxidation-reduction potential. When methylene blue, having a low potential, was used, the reducing agent was captured entirely by the oxygen. No reduction of the dye occurred till all the oxygen was used up (Fig. 6). If, however, a dye of higher potential (e.g., dichlorophenol-indophenol) was used, reduction occurred even in the presence of oxygen, though to a somewhat lesser extent than in its absence. If methylene blue was irradiated in the absence of oxygen and completely reduced, the diffusion of oxygen into the cell resulted in a rapid re-oxidation of the dye. This did not occur when oxygen remained excluded. These reactions are presumably due to the establishment of the reversible equilibria of the types

$$(\text{Dye})^- + O_2 \rightleftharpoons \text{Dye} + O_2^-$$

or

$$(\text{Dye})\text{H} + O_2 \rightleftharpoons \text{Dye} + HO_2.$$

between the reduced dye, or its half-reduced semiquinone form, and oxygen or any other acceptor for the reducing agent which may be present. In addition, the reduced form of the dye is formed nonuniformly throughout the volume according to the dose distribution pattern. This results in a potential difference, which is apparently only very slowly equalized by diffusion of the dye molecules. There are specific qualitative differences in this process between various dyes, apparently due to their different diffusion velocities. Methylene blue was preferred by us in some cases because of its apparently slower diffusion.

While these reversible competitions for the reducing agent could be observed in the system investigated, it appeared that oxidizing reactions due to the hydroxyl radicals could also occur in the same systems. In a gel containing 1 % benzoate but no dye, the formation of phenolic acids could be observed, while when dye was added to this gel before irradiation, the reduction of the dye was greater than in the absence of benzoate. This, of course, is due to the removal of oxidizing OH radicals by the benzoate (Fig. 7).

A complete formal similarity exists between the chemical reactions in aqueous liquids and in aqueous gel systems. Owing to the competition between oxygen and the dyes hitherto investigated, the reactions had to be carried out in the absence of air when

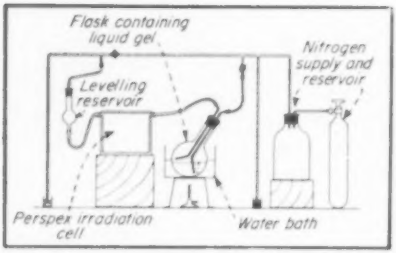


FIG. 8. Apparatus for filling cell with hot gel in atmosphere of nitrogen to exclude air

FIG. 9. Picture of X-ray beam obtained with gel whose calibration curve is given in Fig. 7

quantitative dosimetry was the aim. An apparatus for accomplishing this is shown in Fig. 8.

The curves in Fig. 6 show the effect of irradiating two dyes, in the presence and in the absence of oxygen respectively, demonstrating clearly the competition between the dye and oxygen and also the marked difference in this respect between the two dyes. Because of its greater sensitivity to radiation, and its apparently slower diffusion, we have used a gel of the following composition:

Agar	1 %
Sodium benzoate	1 %
Benzoic acid	0.2 %
Methylene blue	0.003 %

The picture (Fig. 9) shows the result obtained when this gel, contained in a flat Perspex cell, was placed in an X-ray beam. The divergence and attenuation of the beam are clearly seen. It is believed that these experiments may lead to a method of studying dose distributions of wide applicability.

The fact that very small concentrations of suitable acceptors gave reduction yields of the order of unity per ion-pair, even in the presence of large concentrations of agar or gelatine, may be of considerable biological significance. It may indicate the possibility of one particular biological component being selectively affected through a mechanism involving reversible processes in a system composed of many substances. If this were so, it might throw some light on the problem of the remarkable radiosensitivity of some living systems.

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High-Energy Neutron Threshold Detectors (*Continued from page 33*)

tically scattered neutrons are removed only in the latter case.) The ambiguity over which correction factor would be used is not important if the correction is small, as is very often the case.

2. Center-of-mass solid angle. This correction is discussed in reference *9*.

3. Effect of the energy spectrum. This is discussed in reference *8*.

Monitoring. It is often important to know the relative fluxes of fast neutrons for several different accelerator runs, or at different places during the same run. For such purposes, if the neutron spectrum is assumed constant (the usual case), many of the complicating factors discussed under "Procedures" can be eliminated. As is evident from that discussion, if all detectors are of the same material and the same mass and cross-sectional area (the mass is not important for thick detectors), the neutron flux is

$$F = \frac{C\,dN/dt}{1 - \exp(-\lambda\tau)} \qquad (10)$$

where dN/dt is the counting rate at any given length of time after the end of the bombardment, and C is the same constant for all runs and for all positions during the same run. Perhaps the best example of this application was the use of carbon detectors by Sherr (*10*) for the measurement of total cross sections of various materials for fast neutrons.

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* References *1* and *8* are excerpts from reference *11*.

Some Aspects of the BEHAVIOR OF CARRIER-FREE TRACERS—I*

The key to carrier-free-tracer chemistry is the high sensitivity of energetic-radiation detection equipment that makes it possible to estimate extremely small amounts of radioelements. Various methods of coprecipitation, important in this branch of "hot-atom" chemistry, are discussed

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IN THE PREPARATION of radioactive atoms for studies of their behavior in the absence of detectable amounts of isotopic carrier, an attempt is made to exclude all isotopic carrier; but only if stable and long-lived isotopes of the element are nonexistent can it be certain that the radioactive atoms are truly carrier-free. However, it is customary to refer to all radioactive preparations to which no isotopic carrier has been intentionally added and containing no isotopic material detectable by ordinary chemical or spectrographic means as "carrier-free."

The behavior of carrier-free tracers is of interest because the amounts of material involved are so extremely small. For example, the amounts of several radioactive nuclides undergoing 10,000 disintegrations per minute, an easily measurable rate,† are:

1590-year Ra (Ra^{226})
2×10^{-11} gram-atom
140-day Po (Po^{210})
5×10^{-15} gram-atom
10.6-hour ThB (Pb^{212})
1.5×10^{-17} gram-atom
3.1-minute ThC'' (Tl^{208})
7.5×10^{-20} gram-atom (45,000 atoms)

Even if the thorium B and thorium C'' preparations were accidentally contaminated with a million times as many inactive lead and thallium atoms as active atoms, the total amounts of lead and thallium would still be very small. These minute amounts of material studied in carrier-free tracer experiments are referred to as *tracer amounts*. Concentrations of these amounts are referred to as *tracer concentrations*.

A knowledge of the chemical behavior of tracer amounts of substances is important in many fields. Several ele-

* The material in this article, of which Part I is the first half, has been taken from Chapter 6 of the forthcoming book "Radioactivity Applied to Chemistry," Arthur C. Wahl, editor, and Norman A. Bonner, assistant editor (John Wiley & Sons, Inc., New York, 1951).

In addition to the material presented here and in Part II to appear in the March NUCLEONICS, the chapter contains sections on deposition on charcoal and ion-exchange resins, radiocolloid formation, solvent extraction, and vaporization. The references given are by no means complete, but are merely those containing data which well illustrate the phenomena under discussion. The book itself also contains an extensive set of tables which summarize all published experimental data that the authors were able to locate by the end of 1949. Due to space limitations, the six tables dealing with the material of Chapter 6 cannot be included in this article.

Seminar notes prepared during the spring of 1947 by Milton Kahn and Jack M. Siegel were of great aid to the authors in the preparation of this chapter, and the authors are indebted to Dr. Siegel for the use of his notes.

† A disintegration rate of 10,000 per minute is easily measured by instruments detecting individual alpha or beta particles (e.g., Geiger-Müller and proportional counters). Instruments measuring saturation ion currents (e.g., electroscopes and ionization chambers) are usually 10^2 to 10^5 times less sensitive.

ments, namely, francium, astatine, polonium, actinium, promethium, radon, berkelium, and californium exist only as short-lived isotopes. A knowledge of their behavior at ordinary concentrations is chiefly dependent on accurate extrapolations from their behavior at tracer concentrations. The field of "hot-atom" chemistry, wherein the chemical behavior and identity of atoms, ions, and molecules produced by nuclear reactions are sought, is obviously dependent on the tracer chemistry of these substances. In addition to these and other applications, tracer chemistry is itself an interesting field of study.

The behavior of substances at tracer concentrations may differ from that at ordinary concentrations, and such differences are frequently predictable. For example, an equilibrium that lies in one direction when both reactants are present at ordinary concentrations may shift so that it lies in the opposite direction when the concentration of one of the reactants is greatly reduced. This can occur if the reactant and product species contain different numbers of isotopic atoms per molecule. Products formed at low concentrations may not be the same as those formed at ordinary concentrations, either because of these equilibrium shifts or because of rate effects.

Frequently, the concentration of the tracer* is so low that addition of a precipitating agent does not cause the solubility product of the tracer compound to be exceeded, or, if it does, the precipitate formed is imperceptible. However, a tracer may be carried by a foreign precipitate (nonisotopic carrier) formed in or added to the tracer solution.

In studies of the behavior of carrier-free tracers, the measured quantity is always the fraction of tracer that is carried, extracted, oxidized, deposited, etc. The amount (moles) of substance is frequently uncertain, even in order of magnitude, because of possible accidental contamination with isotopic carrier. The amount of tracer substance involved in most carrier-free tracer experiments is extremely small. Hence the loss of even minute amounts by reaction with trace impurities or adsorption on the vessel walls, filter paper, or imperceptible suspended particles of dust and silica may represent sizable fractions of the total. Therefore, such losses are troublesome in carrier-free tracer experiments, whereas in macro experiments these minute losses, being a negligible fraction of the material involved, are not even detected.

The behavior of very long-lived tracers (e.g., U^{238} and Th^{232}) is not considered here because radioactively detectable amounts of them can be treated by ordinary chemical procedures.

COPRECIPITATION

The concentration of a tracer is frequently so low that addition of a precipitating agent does not cause the solubility product of the tracer compound to be exceeded. However, if a non-isotopic carrier that is precipitated by the precipitating agent is present in the tracer solution, some of the tracer may be removed from the solution with the precipitate. The fraction of tracer carried is independent of its concentration over a very wide range because

* The term *tracer* is used somewhat loosely in this discussion, the term sometimes referring only to the radioactive atoms and sometimes to the radioactive atoms plus any isotopic inactive atoms that may be accidentally present. When "tracer" is used in connection with a radioactively measured quantity (e.g., measured ratio of tracer to carrier—counts per minute per milligram), it refers to the radioactive atoms only. When "tracer" is used in connection with a chemical term (e.g., concentration of the tracer, tracer substance, tracer element, tracer compound, tracer ion), it refers to all isotopic atoms, active plus inactive. Because of the chemical identity of isotopes, either interpretation is appropriate when reference is made to the fate of the tracer in a reaction. Unless otherwise indicated, all atoms of the tracer element in the same phase are assumed to be in the same chemical form.

the composition of the precipitate is essentially unchanged by the presence of such minute amounts of material.

The term *coprecipitation* as used here implies carrying by a precipitate formed in the presence of the tracer and does not describe the mechanism by which the tracer is carried. Carrying by preformed precipitates is discussed in Part II of this article.

It is usually possible experimentally to differentiate between two general types of carrying processes: (1) incorporation of the tracer into the crystal lattice of the precipitate and (2) adsorption of the tracer on surfaces of the precipitate during or after its formation. When incorporation into the lattice occurs, the ratio of the fraction of tracer to the fraction of carrier removed from solution is relatively independent of precipitating conditions such as the presence or absence of an excess of the precipitant or of highly charged ions. If the tracer is carried by means of an adsorption process, this ratio is very dependent on precipitating conditions. When a tracer is incorporated into the lattice, the distribution of the tracer is continuous, though not necessarily homogeneous; whereas, if the tracer is adsorbed, its distribution is discontinuous, the tracer being distributed over the surface of the crystals or in some cases only on certain planes of the crystal.

O. Hahn (*1*) classifies carrying processes as follows:

(*a*) *Isomorphous Replacement.* The tracer is incorporated into the crystal lattice, presumably isomorphously since macro amounts of the tracer and carrier compounds form isomorphous mixed crystals.

(*b*) *Adsorption.* The tracer is adsorbed on the surface of a precipitate.

(*c*) *Anomalous-Mixed-Crystal Formation.* The tracer is apparently incorporated into the crystal lattice even though macro amounts of the tracer and carrier compounds do not form isomorphous mixed crystals.

(*d*) *"Internal Adsorption."* The tracer is apparently adsorbed on surfaces of growing crystals, the result being a discontinuous incorporation of the tracer in the precipitate.

Processes *a* and *b* are most clearly understood, and Hahn includes *c* and *d* because it is not always possible to assign carrying processes unequivocally to either *a* or *b*. The discussion in this section is presented according to Hahn's classification.

In general, a tracer ion is efficiently carried by an ionic precipitate if: (1) *the tracer ion is isomorphously incorporated into the precipitate, or* (2) *the tracer ion forms a slightly soluble or slightly dissociated compound with the oppositely charged lattice ion and if the precipitate has a large surface with charge opposite to that of the tracer ion* (*i.e., presence of excess of the oppositely charged lattice ion*).

Via Isomorphous Replacement

When macro amounts of the carrier and tracer compounds form isomorphous mixed crystals,* the tracer ions may replace some of the carrier ions of like charge in the crystal lattice and thus be carried. The efficiency of this carrying process depends on a number of factors, including the lattice energies of the tracer and carrier ions in the carrier compound, the hydration energies of the tracer and carrier ions, and the method of precipitation employed. The last is important because the negligible rates of diffusion of ions in crystals at ordinary temperatures prevent the establishment of equilibrium between the interior of the crystal and

* If two salts are isomorphous (i.e., crystallize in the same form with nearly identical angles), it does not necessarily follow that they form mixed crystals. Isomorphism occurs when the relative sizes of two sets of crystal ions and the effects of polarization on their coordination numbers are similar. If in addition the absolute sizes of the two sets of ions are nearly the same, mixed crystals can form.

the solution. Therefore, unless special care is taken to establish equilibrium (e.g., prolonged digestion to allow recrystallization to occur), the distribution of the tracer in the crystal and between the crystal and the solution is determined as each new layer is formed.

Most quantitative studies of carrying by isomorphous replacement have been made by use of fractional crystallization techniques with moderately soluble salts so that conditions could be accurately controlled. These methods of precipitation, the distributions of tracer obtained, and the two limiting laws these distributions appear to approach are discussed below.

Distribution laws for isomorphous incorporation. If somehow equilibrium between the ions in the interior of the precipitate and the ions in solution is established, the distribution of a tracer in the precipitate will be homogeneous, and its distribution between the precipitate and the solution will follow the Berthelot-Nernst distribution law, which applies to the equilibrium partition of a solute between two phases:

$$\frac{\text{concentration of tracer in solid}}{\text{concentration of tracer in solution}} = K \qquad (1)$$

Since the ratios of the tracer to carrier in both the solid and liquid phases are usually determined, Eq. 1 is somewhat cumbersome, requiring corrections for the density of the solid phase and the concentration of the saturated carrier solution. L. Henderson and F. Kracek (*2*) introduced the following expression or the **homogeneous distribution law:**

$$\left(\frac{\text{tracer}}{\text{carrier}}\right)_{\text{solid}} = \mathbf{D}\left(\frac{\text{tracer}}{\text{carrier}}\right)_{\text{solution}} \qquad (2)$$

D, called the *homogeneous distribution coefficient*, is related to K by the following expression:

$$\mathbf{D} = K(\text{gm solute per cc of saturated carrier solution})/(\text{density of solid}) \qquad (3)$$

For values of **D** greater than unity, the precipitate is enriched in tracer substance; for values of **D** less than unity, the solution is enriched.

A second condition may be realized in which equilibrium is attained between the ions in solution and the ions on the solid surface as each new crystal layer is formed, but in which the recrystallization and solid-diffusion effects are negligible, the ions in the interior of the precipitate retaining their relative positions once covered. This condition results in an inhomogeneous distribution of tracer in the precipitate (concentration near the centers or near the surfaces of the crystals) and in a distribution of tracer between the precipitate and solution that follows the **logarithmic distribution law** derived by H. Doerner and W. Hoskins (*3*). Let x and y be the amounts of tracer and carrier precipitated, and let a and b be the total amounts of tracer and carrier. Then since the amounts dx and dy of tracer and carrier deposited in a given crystal layer are proportional to their respective concentrations,

$$\frac{dx}{dy} = \boldsymbol{\lambda}\,\frac{(a-x)/V}{(b-y)/V} \qquad (4)$$

where V is the volume of the mother liquor and $\boldsymbol{\lambda}$ is a constant characteristic of the system and is called the *logarithmic distribution coefficient.* Integration gives

$$\log\frac{a}{a-x} = \boldsymbol{\lambda}\log\frac{b}{b-y}$$

or

$$\log\frac{\text{total tracer}}{\text{tracer in solution}} = \boldsymbol{\lambda}\log\frac{\text{total carrier}}{\text{carrier in solution}} \qquad (5)$$

For values of $\boldsymbol{\lambda}$ greater than unity, the precipitate is enriched in tracer, which is concentrated near the centers of the crystals; for values of $\boldsymbol{\lambda}$ less than unity, the solution is enriched in tracer, the tracer in the precipitate being concentrated near the surfaces of the crystals.

The similarity between the homogeneous distribution law, Eq. 2, and

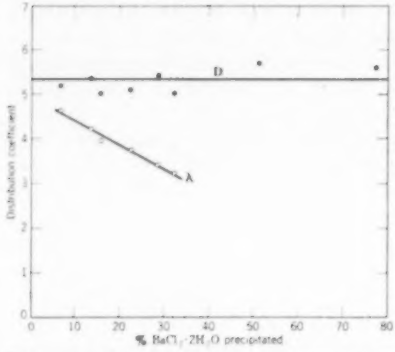

FIG. 1. Distribution of radium after precipitation of barium chloride from supersaturated solutions by vigorous agitation for six hours at 0° C. [V. Chlopin, A. Polessitsky (*5*)]

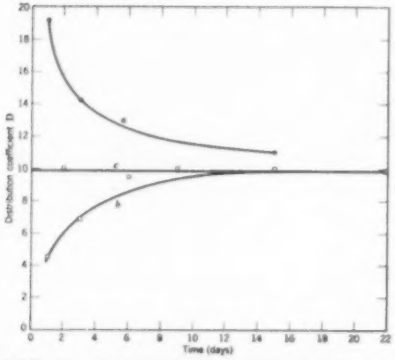

FIG. 2. Approach to equilibrium in the radium-barium bromide system as a function of time; 35° C. Curve *a*: activity initially in solid. Curve *b*: activity initially in solution. Curve *c*: entire system initially in liquid state. [V. Chlopin (*9*)]

the differential form of the logarithmic distribution law, Eq. 4, suggests that, under suitable conditions, **D** and **λ** should have essentially the same values (see Table 2 on page 54).

Rapid precipitation and prolonged digestion—agitated supersaturated solution. Experiments carried out by V. Chlopin and co-workers (*4*, *5*, *6*, *7*, *8*, *9*) showed very definitely that, if a supersaturated solution of a barium salt containing tracer amounts of radium (Ra^{226}) is stirred vigorously at constant temperature for approximately 6 hours, the homogeneous distribution law is followed. Results of experiments carried out with the radium-barium chloride system are shown in Fig. 1, where **D** and **λ** are plotted against the amount of barium chloride precipitated. Solutions supersaturated to different degrees were prepared, and supersaturation completely eliminated by prolonged, vigorous agitation. The homogeneous distribution of radium is indicated by the constancy of **D**.

Since the Berthelot-Nernst distribution law pertains to a system at equilibrium, it was a matter of some importance to establish that the value of **D** is independent of the path of approach to equilibrium. Chlopin (*9*) allowed the radium-barium chloride and radium-barium bromide systems to approach equilibrium in three ways.

(1) The powdered salt containing tracer amounts of radium was added to an inactive saturated solution of the salt, and the mixture vigorously stirred. After various lengths of time, samples of the solution were withdrawn, and the values of **D** determined.*

(2) The method used was the same as the preceding, except that the solution rather than the solid salt had been inoculated with the radium tracer.

(3) A supersaturated solution was prepared by heating the entire system (powdered salt, saturated solution, and radium tracer) until a clear solution was formed and then cooling to the thermostat temperature. The radium-containing barium salt was precipitated by vigorous agitation, which was continued for various lengths of time.

The experiments were carried out at 35° C. In both systems the value of **D** obtained was independent of the method of approach to equilibrium. The results for the radium-barium bromide system are shown in Fig. 2.

* Chlopin used *K* instead of **D**.

The value of **D** for a given system may depend on the temperature. The variation of **D** with temperature for several systems is shown in Fig. 3. If a salt is capable of forming hydrates, an abrupt change in the value of **D** may occur with a slight change in temperature because the composition of the solid phase in equilibrium with the mother liquor changes (see Fig. 3).

Experiments by V. Chlopin and A. Polessitsky (*7*) with the three-component system radium-radium D(Pb^{210})-barium nitrate showed that the two tracers distributed themselves throughout the crystals as though each were present alone. When the system radium-lead-barium nitrate was studied, it was found that for a given ratio of lead nitrate to barium nitrate in the mixed-crystal solid phase, the radium was distributed according to the homogeneous distribution law. When the ratio of lead nitrate to barium nitrate was varied, the value of **D** varied continuously from that found for pure lead nitrate to that found for pure barium nitrate.

Slow precipitation—undisturbed supersaturated solution. R. Mumbrauer (*11*) carried out a series of experiments in which he allowed a number of barium chloride solutions supersaturated to different degrees and containing tracer amounts of radium to stand undisturbed at 0° C. Crystals of barium chloride formed slowly. After several days, when supersaturation had been completely eliminated, the precipitates were separated from their mother liquors and analyzed for radium. The results are shown in Fig. 4, where **D** and λ are plotted against the per cent barium chloride precipitated. The constancy of **D** indicates the applicability of the homogeneous distribution law, the value of **D** being independent of the initial degree of supersaturation.

Mumbrauer (*11*) confirmed the homogeneous distribution of radium in the

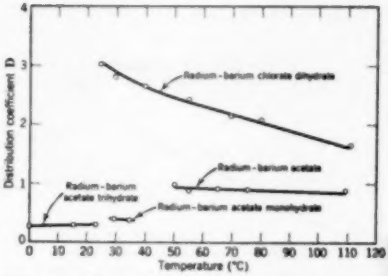
Distribution coefficient D
Radium - barium chloride dihydrate
Radium - barium acetate
Radium - barium acetate trihydrate
Radium - barium acetate monohydrate
Temperature (°C)

FIG. 3. D as a function of temperature for several systems. [B. Goldschmidt (*10*)]

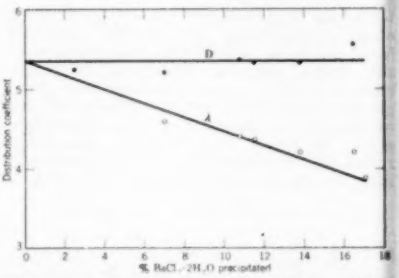

FIG. 4. Distribution of radium after slow precipitation of barium chloride from supersaturated solutions standing undisturbed at 0° C for several days. Supersaturation was completely eliminated. [R. Mumbrauer (*11*)]

crystals in the following way. A series of barium chloride solutions, all supersaturated to the same degree and containing tracer amounts of radium, were allowed to stand at 0° C. The slowly forming crystals of barium chloride were removed from the various solutions at different times and analyzed for radium. The results are shown in Fig. 5, where the ratio of radium to barium in the crystals and values of **D** are plotted against the per cent barium chloride precipitated. It is seen that the ratio of radium to barium in the crystals is independent of the amount of barium chloride precipitated; that is, the radium is homogeneously distributed within the crystals.

As long as the mother liquor is supersaturated, the value of **D** depends on

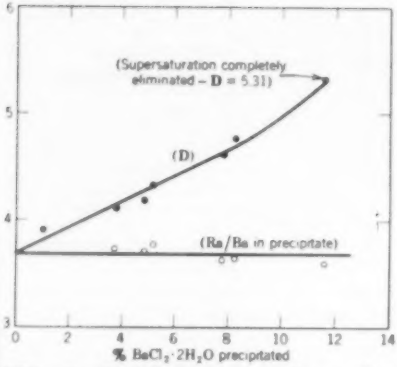

FIG. 5. Distribution of radium after slow precipitation of barium chloride from solutions initially supersaturated to the same degree. Precipitation interrupted before supersaturation was completely eliminated. 0° C. Plotted values of the Ra to Ba ratio in the precipitate were obtained by dividing measured ratios in the precipitates by measured ratio in the initial solution. [R. Mumbrauer (*11*)]

the per cent of barium chloride precipitated. Constant values of **D** (5.37 for the radium-barium chloride system at 0° C), which are independent of the per cent of the carrier precipitated, are obtained only when supersaturation is completely eliminated.

It is interesting to note that the values of **D** obtained for the radium-barium chloride or the radium-barium nitrate systems are the same whether supersaturation is eliminated by vigorous stirring or slow precipitation. The experimental results are shown in Table 1.

The adherence to the homogeneous distribution law of the data obtained from this type of slow precipitation experiment is very surprising and difficult to explain since equilibrium conditions are certainly absent. This phenomenon is discussed by N. Riehl (*12*). Diffusion in the ionic crystals is certainly too slow for the establishment of equilibrium, and N. Riehl and H. Käding (*13*) have shown that the well-formed crystals obtained from this type of precipitation undergo recrystallization only very slowly. Two solutions saturated with barium bromide at 60° C and identical in every respect except that one contained thorium X (Ra^{224}) were slowly cooled to 0° C. The crystals formed in each solution were transferred to the other's mother liquor. These mixtures were kept at 0° C for 24 hours, and then both phases of each mixture were analyzed for thorium X. It was found that the active crystals had yielded essentially no activity to the inactive mother liquor and that the inactive crystals had taken up essentially no activity from the active mother liquor.

Slow precipitation—evaporation of saturated solution. The conditions prevailing when a precipitation is carried out by slow isothermal evaporation of a saturated solution closely approximate those assumed in the derivation of the logarithmic distribution law, namely, equilibrium between the solution and each newly formed crystal layer and nonequilibrium between the interior of the crystals and the solution. Therefore, it is not surprising that data obtained for this method of precipitation obey the logarithmic distribution law of Doerner and Hoskins (page 49).

N. Riehl and H. Käding (*13*; also see *14*) carried out a series of experiments

TABLE 1
Comparison of Values of D
Supersaturation completely eliminated; 0° C.

	D	
System	*Rapid precipitation—Prolonged agitation*	*Slow precipitation—Undisturbed*
Ra-Ba chloride	5.35 (*5*)	5.37(*11*)
Ra-Ba nitrate	1.88*(*11*)	1.88(*11*)

* V. Chlopin (*6*) obtained a value of 2.31 for D. R. Mumbrauer (*11*) believes that Chlopin's value is too high owing to the lack of equilibrium. Mumbrauer stirred his solution for about 12 h; Chlopin stirred his only about 5 h.

in which they evaporated at 20° C saturated solutions of barium bromide containing tracer amounts of radium. The precipitations were carried out over a period of several days. Crystals of barium bromide were removed from time to time and analyzed for their radium content. The results are shown in Fig. 6, where the values of **D** and **λ** are plotted against the per cent of the total barium bromide precipitated. It is seen that **λ** is independent of the amount of barium bromide precipitated, that is, the distribution of radium is in accordance with the logarithmic distribution law.

The results also show that the value of **λ** depends on the rate of evaporation, an indication that complete equilibrium between the surface layers and the solution is not attained. An increase in the rate of evaporation does not necessarily result in a smaller value of **λ**. For example, R. Mumbrauer (*11*) found the value of **λ** to be 1.25 for the radium-barium nitrate system when the evaporation was carried out slowly at 17° C. At a faster rate of evaporation at the same temperature, he found the value of **λ** to be 1.40.

Rapid precipitation and immediate filtration—agitated supersaturated solution. R. Mumbrauer (*11*) showed that

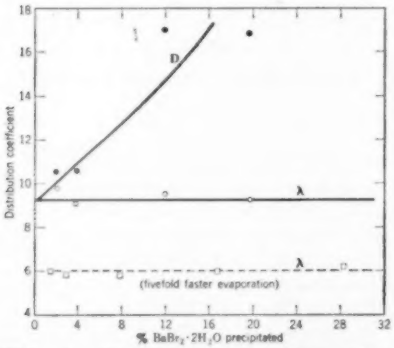

FIG. 6. Distribution of radium after precipitation of barium bromide from saturated solutions by slow evaporation at 20° C. [N. Riehl, H. Käding (*13*)]

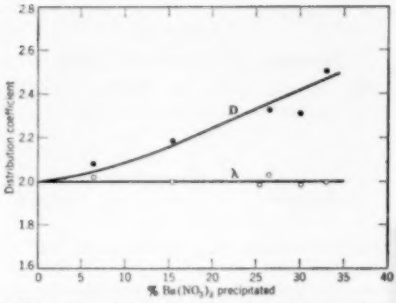

FIG. 7. Distribution of radium after precipitation of barium nitrate from supersaturated solutions by vigorous agitation for a few minutes at 0° C. Precipitate filtered immediately. [R. Mumbrauer (*11*)]

the distribution of a tracer between a precipitate and its mother liquor follows the logarithmic distribution law when the precipitate is formed rapidly by agitation of a supersaturated solution and is immediately filtered. As discussed on page 50, prolonged digestion of such a precipitate before filtration results in the homogeneous distribution, equilibrium being attained through recrystallization.

Mumbrauer investigated this method of crystallization in the following way. Solutions of barium nitrate, each supersaturated to a different degree at 0° C and containing tracer amounts of radium, were vigorously stirred for a few minutes, crystallization being essentially complete in this time. The crystals formed were immediately separated from their mother liquor and analyzed for their radium content. The results are shown in Fig. 7, where **D** and **λ** are plotted against the per cent barium nitrate precipitated. The constancy of **λ**, it being independent of the initial degree of supersaturation, indicates the applicability of the logarithmic distribution law.

The values of **λ** and **D** are compared in Table 2 for the radium-barium chloride and radium-barium nitrate systems at 0° C. The precipitates were

TABLE 2

Comparison of Values of λ and D

Rapid precipitation by vigorous agitation of supersaturated solutions at 0° C.

System	λ *Immediate filtration*	D *Prolonged digestion*
Ra-Ba chloride	4.31(*11*)	5.35(*5*)
Ra-Ba nitrate	2.01(*11*)	1.88(*11*)

formed rapidly by agitation of supersaturated solutions, values of λ being obtained when the precipitates were filtered immediately, and values of D when the precipitates were digested in their mother liquors before filtration.

The expected agreement between λ and D for a given system (see last paragraph on page 49) is fair considering the uncertainty in fulfilling the equilibrium conditions postulated in the derivation of the logarithmic distribution law.

Rapid precipitation after addition of precipitating agent. The degree of local supersaturation caused by addition of a precipitating agent is so erratic that a quantitative study of the type of tracer distribution, like those discussed in the preceding paragraphs, is not practical. However, when isomorphous replacement is possible, carrying by precipitates formed in the presence of the tracer is known to be very reliable, a slight excess or deficiency of the precipitating agent making little difference in the fraction of the tracer carried. For example, O. Hahn, H. Käding, and R. Mumbrauer (*15*, *16*) showed that thorium B (Pb^{212}) forms a type of mixed crystal* with silver chromate and is carried quantitatively by silver chromate when chromate ion is added to a solution containing thorium B and silver ions, regardless of whether the chromate or silver ion is in slight excess.

Possible relationship between solubility and distribution coefficient. It is frequently assumed that in mixed-crystal systems the less soluble compound will concentrate in the solid phase. This is generally true, but there are exceptions such as the radium-barium nitrate system (see Table 3). The data in Table 3 also show that no simple relationship exists between solubility and distribution coefficient. This is not surprising since it is unlikely that the tracer ion will have exactly the same lattice energy in the carrier compound as it has in the pure tracer compound. Also the hydration energies of the tracer ion and the oppositely charged ion in the saturated solution of carrier compound may be somewhat different from those in the saturated solution of tracer compound.

Practical aspects of coprecipitation. Separation of a tracer from a chemically similar carrier (e.g., separation of radium from barium) is of considerable practical importance. Although at the present time the ion-exchange resins appear to offer the most efficient method of separation, fractional crystallization methods have been, and to some extent still are, widely used.

TABLE 3

Solubilities and Values of D at 20° C

[O. Erbacher (*17*)]

System	*Solubility (Moles per 100 gm H_2O)*	*Ratio of solubilities*	D
Ba bromide	0.350	1.91	10
Ra bromide	0.183		
Ba chloride	0.171	2.07	4.5
Ra chloride	0.083		
Ba nitrate	0.033	0.83	1.6
Ra nitrate	0.040		
Ba sulfate	1.06×10^{-6}	1.63	1.8
Ra sulfate	0.65×10^{-6}(*18*)		

* Lead chromate and silver chromate when both are present in macro amounts do not form mixed crystals, but when lead is present in only tracer amounts it is incorporated into silver chromate in a way that resembles mixed-crystal formation (see page 57).

As has been mentioned, the method of crystallization affects the distribution of the tracer and, therefore, the efficiency of the carrying process. A tracer is carried more efficiently the greater the values of **D** and **λ**. The efficiencies for various values of **D** and **λ** are shown in Fig. 8. It is obvious that when **D** and **λ** have the same values, as apparently they usually do have (see Table 2), methods leading to the logarithmic distribution are more efficient than those leading to the homogeneous distribution. For example, if the value of the distribution coefficient is 6, precipitation of 50% of the carrier removes 99.4% of the tracer if it is logarithmically incorporated, but only 86% if it is homogeneously incorporated. Therefore, rapid precipitation from a supersaturated solution followed by the immediate removal of the precipitate from its mother liquor (see page 53) should be the most desirable method because the process is both rapid and efficient. Precipitation by the slow isothermal evaporation of a supersaturated solution, which also results in the efficient logarithmic distribution, is too time consuming to be of great practical importance. In large-scale commercial processes, slow crystallization from a supersaturated solution is most commonly used because less equipment, such as stirrers and filters, is required than for the rapid precipitation process.

Via Adsorption

Even though the tracer and carrier ions are not sufficiently similar for the tracer ion to fit into the lattice of the carrier compound, the tracer may still be efficiently removed from solution by a carrier precipitate through the adsorption process. (The theory of adsorption of ions on ionic precipitates is discussed in Part II of this article.) The following factors affect the efficiency of this type of carrying.

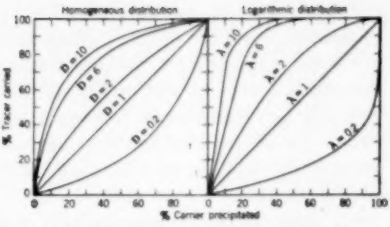

FIG. 8. Efficiency with which the tracer is carried for various values of the distribution coefficients, D and λ

(1) The larger the *surface area* of a precipitate, the larger the fraction of tracer adsorbed. For this reason, a rapidly formed precipitate carries better than a slowly formed one.

(2) As a general rule, the more *insoluble* the compound or the tighter the complex that the tracer ion forms with the oppositely charged lattice ion, the better the tracer ion is carried.

(3) A tracer ion is efficiently adsorbed only if the sign of the *charge* on the precipitate is opposite to its own, i.e., if the oppositely charged lattice ion is in excess. When this is the case, the more highly charged the tracer ion, the better it is adsorbed. The presence of other highly charged ions of the same sign as the tracer ion interferes with the carrying of the tracer.

(4) *Prolonged standing* of the precipitate in contact with the tracer solution may either increase or decrease the fraction of tracer carried. Recrystallization occurring during the standing decreases the surface area, so the fraction of tracer carried will decrease unless the tracer is trapped in the growing crystals (internal adsorption—see page 59), in which case the fraction carried may increase.

The following examples are illustrative of some of the effects mentioned.

O. Hahn, O. Erbacher, and N. Feichtinger (*19*, *20*) studied the coprecipitation of thorium B (Pb^{212}) with calcium sulfate. Because lead sulfate and calcium sulfate do not form mixed

crystals, carrying by isomorphous replacement could not occur. When 48% of the calcium sulfate was slowly crystallized from a hot supersaturated solution, only 1.2% of the thorium B was carried. When the crystallization was repeated from a 10% sulfuric acid solution, 3.8% of the thorium B was carried. However, when the calcium sulfate was rapidly precipitated in the presence of excess sulfate ion by addition of alcohol, the thorium B was efficiently carried. The presence of excess calcium ion interfered with the carrying (see Table 4).

L. Imre (*21*, *22*) studied the coprecipitation of thorium B (Pb^{212}) and mesothorium 2 (Ac^{228}) with silver iodide. When excess iodide was present, both tracers were carried reasonably well (70 to 80%), thorium B being carried by the negatively charged precipitate presumably because of the insolubility of its iodide and because of its $+2$ charge, mesothorium 2 being carried presumably only because of its $+3$ charge. When the silver iodide precipitate was allowed to stand in its mother liquor, the fraction of thorium B carried increased, whereas the fraction of mesothorium 2 carried decreased.

M. Kurbatov, F. Yu, and J. Kurbatov (*23*) investigated the coprecipitation of tracer amounts of 38.8-hour Ba^{133m} with hydrated iron(III) oxide. Assuming that the fraction of tracer barium adsorbed depends on the number of adsorption centers, which these workers assumed to be proportional to the hydroxyl ion concentration, they derived the relation

$$y = a[z(x - x_0)]^b \qquad (6)$$

where y is the fraction of barium adsorbed, z is the quantity of hydrous iron(III) oxide, x_0 is the value of hydroxyl ion concentration above which the solid phase is stable, x is the value of hydroxyl ion concentration, and a and b are constants.

The experimental results obtained under a fair range of conditions support the assumptions made above. When z was kept constant and the OH^- concentration varied from $3 \times 10^{-9} f$ to $3 \times 10^{-6} f$, a value for b of 0.33 was obtained; when the OH^- was kept constant and z varied from 1×10^{-5} to 3×10^{-4} gram-atom a value for b of 0.35 was obtained. Thus Eq. 6 satisfactorily represents the data obtained over a fair range of conditions.

The experiments consisted of titrating a solution containing iron(III), Ba^{133m}, and ammonium chloride to a given *p*H with ammonium hydroxide. The precipitate formed was allowed to settle for a given length of time, portions of the supernatant liquid were removed, and

TABLE 4

Carrying of ThB (Pb^{212}) by $CaSO_4 \cdot 2H_2O$

Calcium sulfate rapidly precipitated in the presence of ThB by addition of alcohol. [O. Hahn, O. Erbacher, and N. Feichtinger (*20*)]

Solution	*$CaSO_4 \cdot 2H_2O$ Precipitated, %*	*ThB Carried, %*	*(% ThB)/(% Ca) Precipitated*
H_2SO_4, 5% excess	91.4	88.0	0.96
H_2SO_4, 10% excess	93.0	85.6	0.92
H_2SO_4, 10% excess	95.8	92.2	0.96
H_2SO_4, 10-fold excess	100	98.4	0.98
$CaCl_2$, 10% excess	85.4	5.2	0.07
$CaCl_2$, 7-fold excess	100	1.7	0.14

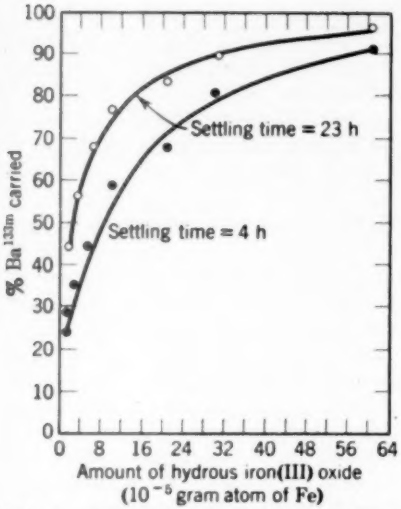

FIG. 9. Effect of amount of hydrous iron (III) oxide on coprecipitation of Ba^{133m}. Conditions: 25° C; pH = 8.01; total Ba = 10^{-11} gram-atom; NH_4Cl concentration = 0.054 f; volume = 32.3–34.4 ml. [M. Kurbatov, F. Yu, J. Kurbatov (*23*)]

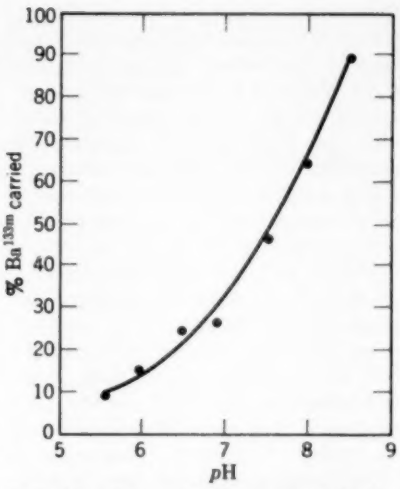

FIG. 10. Effect of pH on coprecipitation of Ba^{133m} with hydrous iron (III) oxide. Conditions: 25° C; total Ba = 10^{-11} gram-atom; Fe = 10^{-4} gram-atom; NH_4Cl concentration = 0.025 f; volume = 13.3 ml; settling time = ~3.25 h. [M. Kurbatov, F. Yu, J. Kurbatov (*23*)]

their Ba^{133m} contents determined. The experimental results are shown in Figs. 9 and 10. It is seen that the amount of barium coprecipitated increases with the amount of iron precipitated, the time of settling, and the OH^- concentration.

M. Kurbatov (*24*) continuing this work, has made a more detailed study of the time effect. He finds that the supernatant solution must be left in contact with the settled hydrous iron(III) oxide for about 40 hours for maximum carrying of Ba^{133m} to occur.

Via Anomalous-Mixed-Crystal Formation

Sometimes a tracer is coprecipitated with a carrier even though conditions for isomorphous replacement and adsorption are absent, i.e., macro amounts of the tracer and carrier compounds do not form isomorphous mixed crystals and the surface area of the precipitate is small. O. Hahn (*1*, *25*) attributes such carrying to *anomalous-mixed-crystal formation* or *"internal adsorption"* and attempts to distinguish between the two processes. Coprecipitation through the "internal adsorption" process is presumably due to the adsorption of tracer ions on surfaces of growing crystals, where the tracer ions are trapped by newly formed crystal layers. This process is discussed in the following section. Coprecipitation through anomalous-mixed-crystal formation is presumably due to the incorporation of the tracer, in some way, into the crystal lattice of the precipitate, anomalous and true mixed crystals composed of tracer and carrier being very similar. Radioautographs of both types of crystals show a continuous distribution of the tracer (*1*). Carrying by incorporation of a tracer in a precipitate to form either anomalous or true mixed crystals is little affected by external conditions (e.g., which lattice ion is in excess or the presence or absence of highly charged foreign ions), which affect carrying due to adsorption. Also

the values of distribution coefficients obtained when anomalous or true mixed crystals are formed are reproducible; those obtained when adsorption is involved are not. The following examples are illustrative of carrying by anomalous-mixed-crystal formation.

Silver chromate, when formed slowly, would not be expected to carry radium or thorium B (Pb^{212}) since it is not isomorphous with barium chromate or lead chromate. O. Hahn, H. Käding, and R. Mumbrauer (*15, 16, 26*) showed that silver chromate did not carry radium but did carry thorium B during slow crystallization from 0.02 *f* nitric acid solutions. The carrying of thorium B resembled carrying by isomorphous replacement, a constant value of **D** = 1.4 being obtained. Also, thorium B is carried quantitatively by silver chromate when chromate ion is added to a solution of silver and thorium B ions regardless of whether the chromate or silver ion is in slight excess. Since the coprecipitation of thorium B with silver chromate resembles the formation of mixed crystals, and since from their macro behaviors these substances are not expected to form mixed crystals, silver chromate containing thorium B is called an anomalous mixed crystal.

It has been known for some time that 26.8-minute radium B (Pb^{214}) and 22-year radium D (Pb^{210}) are concentrated along with radium in the solid phase when barium chloride is precipitated but not when barium bromide is precipitated (*1, 25*). The barium salts crystallize from aqueous solution as the dihydrates, barium chloride in the rhombic system and barium bromide in the monoclinic system; both lead chloride and lead bromide crystallize from aqueous solution as the anhydrous salts in the rhombic system. In spite of their crystallographic differences, barium chloride dihydrate and tracer amounts of lead chloride seem to form a type of mixed crystal, an anomalous mixed crystal. At 0° C the distribution coefficient **D** is 19.6 (*27*). Experiments with added inactive lead carrier indicate the limit of the ability of the barium chloride dihydrate lattice to incorporate lead is reached at about 0.1 mole per cent lead (*1*).

Investigations by H. Käding (*28*) showed that sodium chloride, potassium chloride, and potassium bromide form anomalous mixed crystals with thorium B (Pb^{212}). Here, again, isomorphous replacement is not expected since the crystal structures of lead chloride and lead bromide differ from the sodium chloride type structure. The incorporation of thorium B in the sodium chloride lattice is very pronounced, the value of **D** being approximately sixty for the three salts mentioned above.

The salts were crystallized at 0° C from their supersaturated solutions in the presence of thorium B. Since the halide ion concentration was very high, it is quite possible that some halide complex of lead, such as $PbCl_4^{=}$ or $PbCl_3^{-}$, was incorporated rather than Pb^{++}. Thorium B is not incorporated in potassium chloride if the crystallization takes place from a 1 *f* potassium hydroxide solution, where lead exists as the plumbite ion. Thorium B is carried by silver chloride, which has the sodium chloride type structure, when crystallization occurs from a concentrated solution of hydrochloric acid, but not when it occurs from a 25% ammonia solution. Further evidence for a chloride complex of lead is that chloride ion interferes with the isomorphous incorporation of thorium B in strontium sulfate.

The cesium chloride lattice does not incorporate thorium B. In this respect it is interesting to note that ammonium iodide incorporates thorium B (**D** = 13 at 20° C) only if the crystallization occurs above −17.5° C, where ammonium iodide crystallizes in the sodium chloride type lattice. Below −17.5° C,

where ammonium iodide crystallizes in the cesium chloride type lattice, coprecipitation does not occur.

Via "Internal Adsorption"

Carrying by "internal adsorption" is presumably due to the adsorption of the tracer on surfaces of a growing crystal where it is trapped by subsequently formed layers. It seems likely that the tracer is tightly adsorbed in the rigid layer because of strong attractive forces, which result in insoluble compounds and stable complexes; otherwise the adsorbed tracer would be pushed in front of the newly formed crystal layers rather than being trapped by them. Therefore, tracers that form insoluble compounds or stable complexes with the oppositely charged lattice ion are probably most efficiently "internally adsorbed." However, this condition is apparently not sufficient for the occurrence of "internal adsorption."

"Internal adsorption" is usually characterized by: (1) variations in the values of the distribution coefficient obtained under similar conditions, (2) suppression of carrying by the presence of an excess of the lattice ion whose charge is of the same sign as the tracer or by the presence of other, highly charged ions of the same sign, and (3) ordered discontinuities in the distribution of the tracer in the crystal as shown by radioautographs of well-formed crystals (*1*).

O. Hahn (*1*, *25*) has investigated in detail the incorporation of thorium B (Pb^{212}), thorium X (Ra^{224}), and polonium (Po^{210}) in potassium sulfate, rubidium sulfate, and ammonium sulfate. He found that thorium B and thorium X are strongly concentrated in potassium sulfate and rubidium sulfate (**D**'s = ~20) and are somewhat concentrated in ammonium sulfate (**D** = ~3). Polonium is also incorporated in all three sulfates, but with little concentration (**D** = ~1). On the basis of the following observations, Hahn concluded that the tracers are incorporated by means of "internal adsorption":

(1) The values of **D** varied; for example, those for the incorporation of

(Text continued on page 75)

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Part II, the concluding half of this article, will cover the following subjects: deposition on preformed, non-metallic solids; oxidation-reduction; diffusion and electrical migration; and extrapolation from tracer to macro behavior. See next month's NUCLEONICS

Buildings at the University of Chicago housing the laboratories of the Institute for the Study of Metals, the Institute for Nuclear Studies, and the Institute of Radiobiology and Biophysics appear at the left in the picture above. The accelerator building is in the background, and on the right can be seen one of the towers of the West Stands of Stagg Field where the first controlled nuclear chain reaction was produced

Institutes of the University of Chicago

At the University of Chicago three institutes for advanced research recently moved into a $7.5 million home. These buildings are only part of what Chicago has provided since 1945 in the interests of better facilities for basic research

THE Institute for Nuclear Studies, the Institute for the Study of Metals, and the Institute of Radiobiology and Biophysics were founded at the University of Chicago to continue work on problems growing out of the wartime work of the Manhattan District.

The university built and equipped the Institutes, but industrial organizations pay membership fees to help support their work. These fees—$20,000 per year for membership in one institute, $50,000 for all three Institutes—give companies the opportunity to sponsor long-range programs which none of them could afford individually. Recognizing that tomorrow's technology must come from today's basic discoveries, twenty-seven industrial concerns have joined the Institutes. Industrial members have rights to all publications of the Institutes, royalty-free licenses on any patents taken out and the intangible, but very real, benefits of

quarterly meetings of their scientists with the scientists on the Institutes' staff.

Here three faculty members survey some of the work in progress in their respective divisions.

Institute for Nuclear Studies . . .

By W. F. LIBBY

Like the other University of Chicago institutes, the Institute for Nuclear Studies of which Dr. S. K. Allison is director, is devoted to research and post-doctoral instruction.

The major facilities of the Institute include a 170-inch synchrocyclotron for the production of 450-Mev protons and deuterons and alpha particles of correspondingly lower energies, a 100-Mev betatron, constructed by the General Electric Company, and a 36-inch cyclotron producing 8-Mev deuterons.

The two larger machines are housed in the accelerator building adjoining the Institute's laboratory buildings. The accelerator building includes workrooms and offices for the staffs operating the machines and a few laboratories for research uses. Its design was determined by a track for a heavy crane which runs the length of the building. The crane is used for moving heavy shielding blocks and equipment. The large synchrocyclotron is mounted in a deep pit so that the top of the magnet yoke is at ground level for shielding purposes. The betatron is mounted at ground level and surrounded by a thick shield, together with soundproofing to reduce the noise level.

The new laboratory is a four-story structure linked to two similar buildings for the Institute for Metals and the Institute of Radiobiology and Biophysics. It is of standard design and is large enough to house the staff and a certain number of postdoctoral research men. In addition to the large pieces of accelerator equipment described above, the usual laboratory apparatus will be available, including a 600-kev Cockcroft-Walton accelerator for protons, deuterons, and alpha particles.

The researches in the Institute have been very broad in scope. A very considerable amount of work has been done in geochemistry and cosmology. H. S. Brown has intensively pursued a program of collecting data on elemental and isotopic abundances, principally for meteoric analyses, and the regularities the data have revealed have excited widespread interest. Harold Urey's work on paleotemperatures using the oxygen-18 abundance of calcium carbonate in fossil shells, and his various theories of the origin of the earth and of the composition and characteristics of the moon, have been excellent source materials for many interesting and prolonged discussions. The work on natural radiocarbon and its application to dating has fallen into the same pattern and the methods and results have been of considerable interest to the members of the Institute. A number of distinguished visitors to the University have spoken and partaken in seminars on the general application of nuclear physics and isotopes to cosmology, geochemistry and geophysics.

The more orthodox fields, nuclear physics and radiochemistry, are under intensive investigation. The cosmic ray group have been particularly active in the investigation of the neutron producing component, John Simpson attempting to clearly delineate its variations in intensity with latitude and time, and also attempting to test whether it is similar in these respects to the star-producing component. Marcel Schein has been concerned with almost

every phase of cosmic ray investigation. His recent interest in the heavy primaries has been marked.

During the first four years of the Institute's life, the only accelerators available were the 8-Mev cyclotron and the 400-kev Cockcroft-Walton circuit. Consequently, experimental work on ultra-high-energy nuclear physics is just beginning. With the betatron now in operation and the large synchrocyclotron about to produce a beam, an extensive experimental program is now getting underway. It seems likely that this part of the Institute's interest will grow very rapidly in the near future.

Among the sets of results already obtained, one of the most interesting is from Nathan Sugarman's work on fission of bismuth with the 87-Mev gamma radiation from the betatron. He has demonstrated that fission does occur, and a number of the features of the process such as the cross section and the fission yield curve are in process of determination and measurement. In addition, he has demonstrated the existence of extensive spallation reactions in which neutron emission appears to be favored.

The radioactivities of the ordinary elements is under investigation, two new ones having been found—the isotope rhenium-187 (low-energy negative beta emitter of 10^{12} years' life), and the isotope indium-115 (negative 600-kev beta ray emitter of 10^{14} years' life). The other isobaric pairs are under investigation and two groups are working on double beta emission, the one by direct counting methods and the other under Mark Inghram's direction with the mass spectrometer.

Theoretical physics in the Institute has not suffered for lack of equipment, and as a consequence has forged ahead even in the early stages of the Institute's life. An extensive series of papers has been published on most of the aspects of theoretical nuclear physics and a separate weekly seminar is maintained in this field alone.

The chemical work in the Institute ranges from the purely chemical investigations of Mr. Mayer through tracer applications of isotopes, hot-atom chemistry, and fission-product chemistry, to determinations of nuclear physical data such as half-lives and cross sections, which may require a certain chemical artistry or technique.

The activities of the Institute are characterized by free interchange of ideas and a continuing series of discussions among the members. This phase of the Institute life is brought to a head in the weekly seminars, which the chairman, J. E. Mayer, operates on the principle that the speaker should never be informed before the meeting. At each meeting the chairman asks someone to speak, and this usually serves to catalyze a free-wheeling discussion which may often exceed in length the remarks of the speaker. Speakers have been known to adopt the habit of speaking a few words and then being seated for a few minutes until the discussion dies down, and then adding a few more words, and proceeding in the same manner until their remarks are completed. It is rare indeed that any speaker who begins with the statement that he will take only a few minutes fails to occupy the full hour and a half time of the meeting. The seminars meet weekly throughout the year—there are usually sufficient members in residence even in the summer quarter to justify this.

One of the most fortunate aspects of the Institute's structure is the easy interchange of ideas and suggestions between the physicists and the chemists. In so many universities this does not exist for various reasons, not the least of which is the fact that the two groups are in separate buildings and departments. The organization of the Institute has erased these barriers in this case, and this appears to be bearing fruit.

It is hoped that research men interested in working in the Institute for various limited periods will make their wishes known to the Director or the appropriate members, so that all possibilities can be explored in each case. A definite policy of maintaining a core of visiting research men, particularly in the postdoctoral group, though not limited to this category, exists.

Institute for the Study of Metals . . .

By CYRIL STANLEY SMITH, *Director*

Though formed at the same time as the other Institutes, and though the staff was largely associated with the Manhattan District during World War II, the Institute for the Study of Metals is not a "nuclear" institute, but concerns itself broadly with the structure and properties of metals. The Institute has equipment for ordinary metallurgical studies, and in addition well equipped low-temperature and high-pressure laboratories for extending the range of environments in which the behavior of metals can be studied by the staff.

One of the more important fields of study is that of diffusion, particularly of elements that go into interstitial rather than substitutional positions in a parent lattice. The mechanism of diffusion can be studied simply by the use of a torsion pendulum apparatus to measure internal friction—a device of great simplicity, which has been used to good effect in many diverse investigations. The diffusion of carbon, for example, has been studied from −50° C to 900° C, and found to follow the same rate law through 18 powers of 10. The properties of grain boundaries in solids have been studied partly mechanically, by observing the friction of one grain as it slides over another, and partly in terms of the interface energy (surface tension) of the boundaries, for it has become apparent that the microstructure of annealed metals and alloys is largely dependent on surface tension. A wide variety of such interfaces is now being studied, and the requirements of interface energy and of space-filling are being united.

In the low-temperature field, studies are in progress on the brittleness of pure iron and various iron alloys at low temperatures, on the electrical and magnetic properties of superconductors, and on the properties of liquid helium II. The latter (hardly a metal!) has been shown to possess superfluidity even in adsorbed films only two atoms thick. The association of the low temperature and metallurgical groups in our laboratories has resulted in the discovery of two new transformations in metals—in lithium and in indium-thallium alloys. Both of these transformations are of the diffusionless type and their study is expected to throw light on the nature of the similar but more complicated transformation that occurs in steel and is responsible for its hardness.

Reactions that occur at the surface of metals are of great importance, both in high-temperature service and in corrosive environments. Oxidation and reduction have been studied by the use of a microbalance in single crystals of various orientations, and electron diffraction studies of the nature of the condensed film of various substances on a crystalline substrate. Attempts have been made to study electronically the nature of passivity in corrosion media.

Radioactive tracers are being used to study self-diffusion in alkali metals and will be extended to diffusion in dilute solutions of other systems. The defects

that may be involved in the diffusion mechanism are difficult to study; consequently studies are also being done on ionic crystals, particularly the silver halides. Other work on non-metallic crystals is that on ferroelectrics.

Institute of Radiobiology and Biophysics . . .

By THORFIN R. HOGNESS, *Director*

The Institute of Radiobiology and Biophysics was organized in 1945 to provide a continuity of the work on the biological effects of radiation carried out during the last war, and to help provide a fundamental basis for the interpretation of the vast amount of data accumulated at that time.

The field of radiobiology was given an impetus by atomic energy. As the experimenters in this field delved more and more into the fundamental aspects of radiobiology, the more they were carried away from the immediate field of the effect of radiation on the complex living organism. To understand the whole they were forced to begin with the part, the smallest unit of which is the cell. So, in the course of five years, the work on the effects of radiation has become largely confined to the action of radiation on the individual cell.

At the present time the University of Chicago undertakes to train graduate students in biophysics, but none of those in charge of this course are able to give a definition of biophysics or even to limit its scope. We therefore first make sure that the student has a very thorough knowledge of the physical sciences and then we turn him loose in a biological field. Odd and seemingly unrelated pieces of research may therefore be expected to emerge from the Institute of Radiobiology and Biophysics.

One of the problems with which our scientists are concerned is: By what microscopic mechanism does radiation kill a cell? The problem of killing by radiation probably is intimately tied to the problem of mutation.

What is the effect of radiation impinging directly on the chromosomes as compared with that impinging only on the cytoplasm? Will the mutation rate and killing be the same in the two cases? The answer to these questions is being sought by R. E. Zirkle, W. Bloom, and their collaborators.

The problem is not a simple one, for the cells to be worked with are very small. The experiment is so designed that a very narrow beam (about one micron in each cross-sectional dimension) of high-energy protons can be directed on a small cross section of the cell. Protons of 2 Mev energy are needed to penetrate the cell. For this purpose, a $100,000 Van de Graaff generator was built.

In the meantime, Zirkle, Bloom, and their co-workers carried out preliminary experiments on the whole cell with simpler forms of apparatus.

At the instigation of Paul C. Hodges, head of the Section on Radiology at the University of Chicago, Robert J. Moon has developed a new type of X-ray apparatus. This apparatus employs a scanning X-ray tube and a scintillation counter; the X-ray image is seen on the screen of a kinescope.

The advantages of such a tube are manifold. (1) The roentgenologist will be able to see the fluoroscopic image in broad daylight if necessary. (2) Scattered radiation will not produce fuzzy edges of the image. (3) By the use of pulse-height selector tubes, scintillations of any range of magnitude can be selected for recording. (4) Memory tubes also to be installed will retain any

picture. (5) Motion pictures of the subject may be taken. (6) Since the concentration of X-rays passing through the subject will be about 1/100 that of present practice, the orthopedist may eventually be able to set broken bones while observing the patient on the kinescope.

Another of Dr. Moon's developments is a very fast counting circuit—10^7 counts per second are now possible. He is also developing a fast neutron spectrograph which involves counting scintillations produced by neutrons and screening these scintillations by means of pulse-height selector tubes.

Leo Szilard, a physicist, and A. Novick, a chemist, have in the last few years become microbial geneticists. In their studies on the mutations of bacteria they have invented the "chemostat," which is extremely valuable in growing bacteria under controlled conditions.

With this device Szilard and Novick have studied the spontaneous mutation rate of one strain of bacteria. Selecting three different conditions such that the generation times of the bacteria were two hours, six hours, and twelve hours, all at the same temperature, they found that the rate of formation of a particular mutant does not depend upon the rate of division of the bacteria but rather is a function of time only. In other words, the process which brings about mutation goes on at the same rate regardless of whether or not the bacteria propagate at a fast or slow rate. These investigators have found also that the reaction causing mutation has an activation energy of about 20,000 calories. This information gives us some little insight into the kind of fundamental process which is taking place.

Robert D. Boche is studying the mutations of algae (microscopic plants) brought about by X-rays. He has made many new strains, some of which have lost their green color. Some information on the fundamental process of photosynthesis may be obtained from these odd forms of algae.

J. Franck, H. Gaffron, and their collaborators are studying the mechanism of photosynthesis; Professor Franck is basically a physicist, having received the Nobel Prize for work in this field. Professor Gaffron is a biochemist.

In the last ten years it has been shown that *free radicals*, unstable and chemically incomplete molecules, play a very important role in a great number of chemical reactions. The formation of high polymers such as lucite and rubber are examples. There is growing evidence that free radicals also take part in biological reactions and are one of the contributing agents bringing about mutations. Morris S. Kharasch of the staff has an international reputation for his work on free radicals.

George H. Marmont is studying the mechanism of nerve action and at odd times the mechanism of electric current production of the electric eel.

Leon O. Jacobson finds that 42 per cent of rats irradiated by a 100 per cent lethal dose of X-rays survive if the spleen is protected by shielding against the rays. The spleen generates the red corpuscles of the blood.

C. Phillip Miller finds that one of the major factors in causing death from radiation is the injury to the wall of the intestine, to the extent that such injury allows the normal intestinal bacteria to penetrate into the blood stream and therefore give rise to what is known as bacteremia.

A description of some of the other important projects, particularly that of Konrad E. Bloch on the biosynthesis of proteins (growth), that of Richard Abrams on nucleic acids (the chemistry of reproduction), and that of J. Garrott Allen on blood clotting, requires too much elaboration to be appropriate for an article of this kind. —END

Line Noise Interference with Particle Counting

Accuracy of low-level counting often is impaired by oscillatory power-line transients. A study of these disturbances shows that they can be reduced but not eliminated. A simple line-noise monitor aids selection of valid counting periods

By C. G. GOSS and F. M. GLASS

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MOMENTARY BURSTS of counts which often occur when a nearby power switch is thrown are well known to research workers using radiation counting equipment. These bursts may come from lightning, from switch operation on any small inductive load such as a soldering gun, or from power switch operation on most electrical equipments, but faulty or blinking fluorescent lights are, perhaps, the most troublesome in this respect. For those doing low-level counting or those counting small pulses with moderate-gain amplifiers, this sort of disturbance may render accurate work impossible.

Investigation of Noise

Experiments have shown that fluorescent lights can cause large noise bursts. For example, when the lights in each dual 40-watt fluorescent fixture flash on, a 20-volt (peak-to-peak) burst of oscillations is produced on the power line for approximately 100 μsec. As observed on the 5 and 25 μsec driven sweeps of the Dumont 248 oscilloscope, the waveforms of the oscillations are very ragged. The fundamental frequency seems to vary from 100 to 300 kc, and there are transitions, spikes, etc., that appear to have appreciable energy distributed over the frequency spectrum to well above one megacycle. A faulty fluorescent tube will flash visibly from one to five times per second and will produce an equal number of bursts of oscillations on the power line from 100 to 5,000 μsec long, plus other smaller, shorter duration bursts. All quoted amplitudes and frequencies were observed with the 60-cycle component of the power line attenuated by means of a high-pass resistance-capacitance filter.

With frequency components in the megacycle range of such amplitude, the transient bursts experience little difficulty in reaching the input grid of the amplifier stages by capacitative coupling either through the circuit wiring or through the power transformer and filament leads. Also, if a preamplifier is used, voltages may develop along the cable shield, which acts as a receiving antenna for radiated noise. These voltages will appear on the grid of the first stage in the main amplifier of the instrument.

Little difference was noted between fluorescent tubes, fixtures, and conventional starters of different manufacturers, or between tubes emitting various colors. But of the two positions a tube may occupy in a fixture, one position will allow about ten times as much disturbance from the tube and starter to reach the power line as the other position. This was attributed to the phase-shifting technique used in the

ballast to lessen the stroboscopic effect commonly observed with single-tube fluorescent units.

Noise Reduction

By equipment design. No simple experimental remedy is presented, but some suggestions can be made. The amplitude and prevalence of these interfering signals emphasize the need for: proper shielding of the amplifier, preamplifier, and detection unit; resistance-capacitance decoupling of the low-level amplifier stages with electrolytic capacitors of low series inductance (a feature not common to all manufacturers' products); careful placement of the various pieces of equipment used in each experiment; and a carefully planned and tested ground system or floating ground. Either multiple grounds, (i.e., amplifier and preamplifier chasses grounded) or long preamplifier cables are particularly bad.

Several companies build a fluorescent light starter that locks itself out of operation after flashing a faulty tube for one to two minutes, but these starters, although helpful, constitute only a partial solution.

By filtering. Filtering of one sort or another is usually attempted in many fields of electronics when difficulties arise from interference. Often the preferred place to filter is at the source of the interference, but there are too many potential sources for this to be a practical solution in this case. Further, the source impedance of the arc is so low that merely hanging a few capacitors on the line at various points produces no noticeable reduction of noise amplitude. Thus, the 20-volt bursts of oscillations are present on all lines that connect to the same terminal on the transformer feeding the faulty light. The attenuation through a distribution transformer has not been measured, but it is expected to be high.

The line could be filtered at the point at which it enters the equipment, but this does not reduce the noise level in the lighting and utility circuits that usually run within a few feet of the equipment. Pickup from these lines can be serious, even in low-impedance circuits and in cable shields. Two 3-foot clip leads on the vertical input and ground terminals of a Dumont 304-H oscilloscope, shorted together at their far ends and lying on a work bench no closer than two feet to a power line, picked up 50 mv peak of noise from the 20-volt oscillations on the power line.

It is the authors' opinion that filtering can be most effective only if it is planned to isolate an area completely. Filtering at the power entrance to an electrically shielded room is ideal. The next best choice is the power entrance (preferably underground) of the building—the smaller the building, the better. Larger buildings might be broken into several different blocks, but it would be more difficult and more expensive to achieve high-frequency isolation if it were not provided for in the original building plans.

Since most research workers do not have facilities that include noise-free power lines and electrically shielded rooms, it is expedient to take a few precautionary measures. Line noise can be attenuated 40 db or more before it enters the instrument power supply by a low-pass filter and sine-wave voltage regulator. Since this only prevents the noise from reaching the instrument through the power supply and does not prevent pickup from radiated noise, this precaution alone is not certain to give noise-free operation. Therefore, some type of monitoring device to indicate when line noise occurs will be helpful.

Line Noise Monitor

The device shown in Fig. 1 can be used either as a monitor in counting

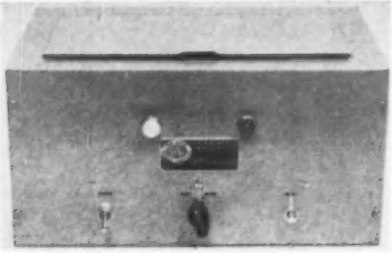

FIG. 1. Line noise monitor

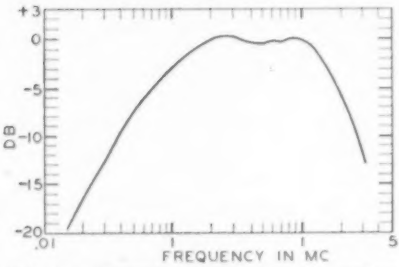

FIG. 2. Frequency response of monitor

rooms to indicate when data should be rejected, or as a building monitor to detect faulty wiring or noisy appliances. It is not a quantitative instrument, and serves only one purpose: that of sounding an alarm or recording the occurrence of noise disturbances. Continuous monitoring makes it possible for the technician to know when line transients introduce an error in counting data and thus to take a recount. Quite often a source of continuous or intermittent noise develops in a building and continues for some time before it is detected and eliminated. When used as a building monitor the instrument may be operated to sound an alarm only when transients persist or recur frequently. This makes it possible to detect the noise when it occurs, without tripping the alarm or recorder on short bursts of noise produced by closing or opening switches on the circuit.

The monitor has been made as simple, compact, and as low in cost as is consistent with good design and dependability. It is housed in a 15 × 7 × 8 in. cabinet, having three controls, including the "on-off" switch, on the front panel. The sensitivity is variable from 0.25 volts to 8 volts (at 1 megacycle). The frequency response to line noise corresponds very closely with that of a linear amplifier when operating on the 2-megacycle band width. Figure 2 shows the frequency response curve of the monitor.

The circuit consists of a one-stage amplifier, a trigger pair, an integrating circuit and a relay stage. The circuit is powered by a conventional power supply employing a VR-150 regulator tube to regulate the plate supply. See Fig. 3.

The signal is taken directly from the line through a high-pass filter. The amplified signal operates the trigger pair, the output of which is rectified and used to charge a capacitor in the grid circuit of the relay tube. The 6SH7 relay tube is normally cut off and conducts only when the integrating capacitor is charged positively. When the 6SH7 conducts, the relay in its plate circuit (RY_1) is energized, closing two sets of normally-open contacts. One contact supplies 110 volts a-c for the operation of an alarm or recorder. The other contact energizes RY_2 which in turn closes two normally-open contacts, one of which is used as a holding contact, while the other energizes a red panel light.

RY_1 drops out as soon as the positive charge leaks off the integrating capacitor, thus de-energizing the alarm or recorder circuit. Since RY_2 sealed itself in, the red light remains on until the reset switch (S_4) is opened. The reset switch also has a normally-open set of contacts which serve the purpose of removing any charge left on the integrating capacitor when the reset switch is operated. The selector switch (S_2) and neon light indicator make it

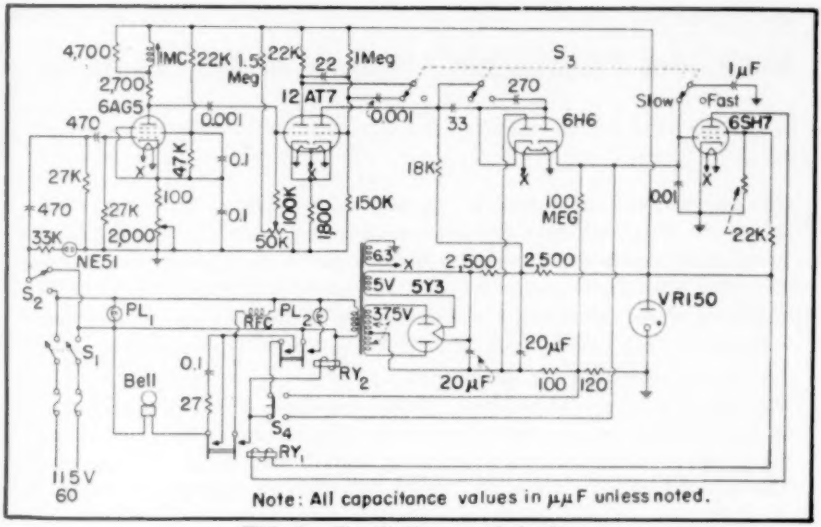

FIG. 3. Circuit diagram of monitor

possible to select the hot side of the line when discrepancies in three-wire systems occur.

The high-pass filter in the input grid circuit and the combination inductive and resistive load in the amplifier plate are chosen to produce the frequency response shown in Fig. 2. This curve refers to over-all trigger sensitivity, and is therefore affected by the frequency response of the trigger pair.

The ability of the monitor to detect either single bursts of noise or, when used as a building monitor, to detect only continuous or frequent noise bursts is accomplished by changing both the resolving time of the trigger pair and the integrating time. When S_3 is in the fast position, the resolving time of the trigger pair is approximately 4 μsec and the capacity of the integrating capacitor is 0.1 μf. The coupling capacitor between the trigger pair and the integrator is 303 μμf. In this position noise of 1 msec duration in the frequency range of 50 kc to 3 Mc will trigger the relay tube.

When S3 is in the slow position, the resolving time of the trigger pair is approximately 120 μsec, and the capacity of the integrating capacitor is 1.1 μf. The series coupling capacitor now has a capacity of 33 μμf, and only a small portion of the current the trigger pair is capable of delivering is integrated. This mode of operation requires a continuous line noise for a period of 300 msec, or from 300 to 350 bursts of 1 msec duration occurring within the time constant of 110 sec, to trigger the relay tube. This is well within the range of a single defective fluorescent light but will not alarm when a whole roomful of fluorescent lights are turned on.

The sensitivity is controlled by a potentiometer in the cathode of the 6AG5 amplifier. The range of 30 in sensitivity has proved adequate for needs here. However, the range can easily be increased if the need arises. It is the intent of the laboratory to place these monitors in all counting rooms in this installation. —END

ICRP supplement on

Maximum Permissible Amounts of Radioisotopes

The International Commission on Radiological Protection (ICRP) finds that, at present, it cannot make firm recommendations regarding the maximum permissible amounts of radioactive isotopes that may be taken into, or retained in, the body. On the basis of the general principles set forth in its recently revised international recommendations on radiological protection (published in the January NUCLEONICS*), it is possible, however, to make reasonable calculations of the maximum permissible amounts of several of the more important radioisotopes. In the meantime, ICRP draws attention to the following data on maximum permissible exposures to radioisotopes for occupational workers, presently used in the United States, Canada, and the United Kingdom.

THE RADIOACTIVE ISOTOPES considered below can enter the body of the occupational worker either by inhalation, or by ingestion. Accordingly, figures are also given for the maximum permissible levels (mpl) of various isotopes in air and in liquid media.

Two appendixes provide quantitative data of related interest. Miscellaneous biological values for *Standard Man* are given in Appendix I, and the values adopted for the *Relative Biological Efficiency* of any given radiation are tabulated in Appendix II.

Radium-226

Clinical observations on chronic radium poisoning in man indicates that the most serious effects are anemia and damage to bone, including osteogenic sarcoma. Both effects appear to have a threshold of the order of 1μc fixed in the skeleton. Accordingly, it is accepted that:

(a) *The maximum permissible amount of radium fixed in the body is 0.1 μc.*

(b) *For radium appearing in the atmosphere as a soluble aerosol, assuming that 25% of the inhaled amount is absorbed and that 25% of the absorbed amount is retained with a mean life of about 10^4 days, the maximum concentration in air for soluble compounds is 8×10^{-12} μc/cc.*

(c) *If radium enters the body through liquid media, assuming 10% of the ingested amount is retained with a mean life of about 10^4 days, the maximum permissible concentration of the liquid is 4×10^{-8} μc/cc.*

* See "Radiological Protection—International Commission Recommendations" [NUCLEONICS 8, No. 1, 31 (1951)] to which this article is a supplement. The International Commission on Radiological Protection met in London in July, 1950, and adopted the new radiation safety standards here referred to, for release on January 1, 1951. For the names of the ICRP members who prepared this supplement, see page 32 of reference article.

Plutonium-239

On the basis of the relative biological effects of plutonium and radium, as observed in animal experiments, it is accepted that:

(a) *The maximum permissible amount of Pu^{239} fixed in the body is 0.04 μc.*

For soluble compounds of plutonium in the atmosphere, it is estimated that 10% of the inhaled material is absorbed, with a mean life of 10^4 days. The maximum permissible concentration in air is, therefore, 2×10^{-12} μc/cc.

For insoluble compounds, it is estimated that the mean life in the lung is 200 days. If the irradiation of the lungs by alpha rays were limited to the biological equivalent of 0.3 r per week, the corresponding concentration of the plutonium in air would be 7.5×10^{-12} μc/cc. In view of the possibility of the transference of some of the insoluble material from the lungs to the skeleton, it is *suggested* that:

(b) *The maximum permissible concentration of Pu^{239} in air is 2×10^{-12} μc/cc, for soluble and insoluble compounds.*

(c) *For Pu^{239} in liquid media, assuming that 0.1% of the ingested amount is retained in the skeleton with a mean life of 10^4 days, the maximum permissible concentration is 1.5×10^{-6} μc/cc.*

Strontium-89 and Strontium-90 (plus Yttrium-90)

On the basis of the observed relative biological effects of Sr^{89} and Ra on animals, it is accepted that:

(a) *The maximum permissible amount of Sr^{89} in the body is 2.0 μc.*

Since the combined disintegration energy of the $Sr^{90} + Y^{90}$ pair is twice that of Sr^{89}, the maximum amount of Sr^{90} which can be permitted in the body is only one-half that of Sr^{89}. Accordingly:

(b) *The maximum permissible amount of Sr^{90} in the body is 1.0 μc.*

If strontium is assumed to behave like radium as regards uptake, then

(c) *For Sr^{90} in air, assuming that 25% of the inhaled amount is absorbed and 25% of the absorbed amount is retained with a mean life of about 15 years, the maximum permissible concentration is 2×10^{-10} μc/cc.*

(d) *For Sr^{90} in liquid media, assuming 10% of the ingested amount is retained with a mean life of about 15 years, the maximum permissible concentration is 8×10^{-7} μc/cc.*

Natural Uranium

As the specific activity of natural uranium is so low, it is considered that the hazards arising from its use are mainly chemical.

Polonium-210

Although polonium is not a bone-seeking isotope, some data exist on its toxicity relative to radium in animals. On this basis, it is accepted that:

(a) *The maximum permissible amount of Po^{210} in the body is 0.005 μc.*

Tritium

It is assumed that tritium (H^3) will be encountered in chemical forms in which free exchange takes place with ordinary hydrogen in the aqueous vapor in the lungs. If the mean energy of the beta radiation is 5.5 kv, a concentration of 0.14 μc per gram of tissue will result in the biological equivalent of 0.3 r per week. The mean life of H^3 in the body is taken to be 10 days. A concentration of 0.14 μc per

gram of tissue corresponds to 10 mc uniformly distributed in the 70 kg of the Standard Man (see Appendix I). It is, therefore, accepted that:

(a) *The maximum permissible amount of H^3 in the body is 10 mc.*

(b) *The maximum concentration of H^3 in air, based on a permissible daily intake of 1 mc and complete absorption in the lungs, is 5×10^{-6} μc/cc.*

(c) *The maximum concentration of H^3 in liquid media, based on a permissible daily intake of 1 mc, and complete absorption in the body, is 0.4 μc/cc.*

Carbon-14 (as carbon dioxide in air)

A rate of energy absorption biologically equivalent to 0.3 r per week would be produced by 0.014 μc of C^{14} per gram of tissue. If the highest proportion of carbon in any tissue is 50%, then the maximum permissible concentration of C^{14} in carbon in the body is 0.028 μc per gram of carbon. The postulated route of entry of C^{14} into the body is via the alveoli of the lungs, and the isotopic concentration in the alveolar air must therefore be limited to 0.028 μc per gram of carbon. Since alveolar air contains 5.5% by volume of carbon dioxide, the maximum permissible concentration of C^{14} in the carbon dioxide of the alveolar air is about 1×10^{-6} μc/cc. Accordingly:

(a) *The maximum permissible concentration of C^{14} as carbon dioxide in air is 1×10^{-6} μc/cc.*

Sodium-24

The energy (beta and gamma) absorbed in the body per disintegration of Na^{24} is estimated to be 2.7 Mev. Since sodium is uniformly distributed throughout the body:

(a) *The maximum permissible amount of Na^{24} in the body, corresponding to a dose rate biologically equivalent to 0.3 r per week, is 15 μc.*

Since biological excretion may be neglected in comparison to the radioactive decay, for which the mean life is 0.8 day:

(b) *For Na^{24} in liquid media, the maximum permissible concentration, assuming complete absorption, is 8×10^{-3} μc/cc.*

Phosphorus-32

Experimental and clinical data in man show that at times of the order of the mean life of P^{32}, i.e., about 20 days, the concentration of P^{32} in red bone marrow reaches a value only about three times the average concentration for the whole body. As most of the P^{32} is still diffused throughout the body, a dose rate biologically equivalent to 0.3 r per week in the critical tissue (red bone marrow) is, therefore, produced by a total quantity of approximately 25 μc of P^{32} in the whole body. To allow for the possible occurrence of higher local concentrations in bone marrow:

(a) *The maximum permissible amount of P^{32} in the body is 10 μc.*

(b) *The maximum permissible concentration of P^{32} in liquid media assuming that 100% is absorbed and that the biological excretion can be neglected in comparison to the radioactive decay is 2×10^{-4} μc/cc*

Cobalt-60

Making the assumption that all the cobalt which is absorbed is deposited in the liver and that the effective energy in the liver is 1.3 Mev per disintegration, the

Maximum Permissible Levels for Occupational Exposure

	MPL in body (μc)	*Effective mean life (days)*	*Permissible daily deposition in body (μc)*	*Proportion absorbed via lungs and retained in body*	*MPL in air (μc/cc)*	*Proportion retained from gut*	*MPL in liquid media (μc/cc)*
Ra^{226}	0.1	10^4	10^{-5}	0.06	8×10^{-12}	0.1	4×10^{-8}
Pu^{239}	0.04	10^4	4×10^{-6}	0.1	2×10^{-12}	0.001	1.5×10^{-4}
Sr^{89}	2.0	....	.....	.....	.....	.....	.....
$Sr^{90}(+Y^{90})$	1.0	5,000	2×10^{-4}	0.06	2×10^{-10}	0.1	8×10^{-7}
Po^{210}	0.005	.....	.....	.....	.....	.....	.....
H^3	1×10^4	10	1×10^3	1	5×10^{-5}	1	0.4
C^{14} (as CO_2 in air)	.....	.....	.....	.....	1×10^{-6}	.....	.....
Na^{24}	15	0.8	20	.....	.....	1	8×10^{-3}
P^{32}	10	20	0.5	.....	.....	1	2×10^{-4}
Co^{60}	1	20	0.05	.....	.....	1	1×10^{-5}
I^{131}	0.3 (0.18 in thyroid)	12	0.015 (to thyroid)	0.2 (to thyroid)	3×10^{-9}	0.2	3×10^{-6}

amount of Co^{60} to give a dose rate biologically equivalent to 0.3 r per week is 1 μc. It is, therefore, accepted that:

(a) *The maximum permissible amount of Co^{60} in the body is 1 μc.*

(b) *The maximum permissible concentration of Co^{60} in liquid media, assuming 100% absorption and a half-life in the liver of 20 days, is 1×10^{-5} μc/cc.*

Iodine-131

The energy absorbed in the thyroid gland per disintegration of I^{131} is estimated to be 0.27 Mev, and so the amount of I^{131} in the gland to give a dose rate biologically equivalent to 0.3 r per week is 0.18 μc, which would correspond to about 0.3 μc in the body. It is, therefore, accepted that:

(a) *The maximum permissible amount of I^{131} in the body is 0.3 μc.*

(b) *The maximum permissible concentration of I^{131} in air, assuming that 100% is absorbed, that 20% of the absorbed amount is deposited in the thyroid, and that biological excretion from the gland can be neglected in comparison to radioactive decay, is 3×10^{-9} μc/cc.*

(c) *The maximum permissible concentration of I^{131} in liquid media, assuming that 100% is absorbed, that 20% of the absorbed amount is deposited in the thyroid gland, and that biological excretion from the gland can be neglected in comparison to radioactive decay, is 3×10^{-5} μc/cc.*

Note: All the preceding permissible amounts of isotopes refer to occupational exposure, and are summarized in the table at the top of this page.

(Continued on the next page)

Appendix I—Standard Man

Mass of Organs:

Organs	*Grams*
Muscles	30,000
Skeleton,	
Bones	7,000
Red Marrow	1,500
Yellow Marrow	1,500
Blood	5,000
Gastro-intestinal tract	2,000
Lungs	1,000
Liver	1,700
Kidney	300
Spleen	150
Pancreas	70
Thyroid	20
Testes	40
Heart	300
Lymphoid tissue	700
Brain	1,500
Spinal cord	30
Bladder	150
Salivary glands	50
Eyes	30
Teeth	20
Prostate	20
Adrenals	20
Thymus	10
Skin and subcutaneous tissues	8,500
Other tissues and organs not separately defined	8,390
Total Body Weight	70,000

Chemical Composition:

Element	*Proportion Per cent*	*Approximate mass in grams in the body*
Oxygen	65.0	45,500
Carbon	18.0	12,600
Hydrogen	10.0	7,000
Nitrogen	3.0	2,100
Calcium	1.5	1,050
Phosphorus	1.0	700
Potassium	0.35	245
Sulphur	0.25	175
Sodium	0.15	105
Chlorine	0.15	105
Magnesium	0.05	35
Iron	0.004	3
Manganese	0.0003	0.2
Copper	0.0002	0.1
Iodine	0.00004	0.03

The figures for a given organ may differ considerably from these averages for the whole body. For example, the nitrogen content of the dividing cells of the basal layer of skin is probably nearer 6% than 3%.

Applied Physiology: Average data for normal activity in a temperate zone.

Water balance—

Daily water intake

In food (including water of oxidation)	1.0 liter
As fluids	1.5 "
	2.5 liters

Calculations of maximum permissible levels for radioactive isotopes in water have been based on the total intake figure of 2.5 liters per day.

Daily water output

Sweat	0.5 liter
From lungs	0.4 "
In feces	0.1 "
Urine	1.5 "
	2.5 liters

Total water content of body 50 liters

Respiration—

Area of respiratory tract

Respiratory interchange area	50 m²
Non-respiratory area (upper tract and trachea to bronchioles)	20 m²
Total	70 m²

Respiratory exchange

	Physical Activity	
	At work	Not at work
Hours per day	8	16
Tidal air (liters)	1.0	0.5
Resp. per minute	20	20
Volume per 8 hours (m³)	10	5
Volume per day (m³)	20	

Carbon dioxide content (by volume) of air

Inhaled air (dry, at sea level)	0.03%
Alveolar air	5.5%
Exhaled air	4.0%

Retention of particulate matter in the lungs—

In those cases where specific data are lacking, the convention has been adopted that 50% of any aerosol reaches the alveoli of the lungs. If the particles are soluble, they have been considered to be completely absorbed; if insoluble, then the 50% amount has been regarded as retained for 24 hours, after which only half of it, i.e., 25% of the inhaled amount, is retained *in situ* indefinitely.

Duration of exposure:

Duration of occupational exposure. The following figures have been adopted in calculations pertaining to occupational exposure:

8 hours per day
40 hours per week
50 weeks per year

Duration of "Lifetime" for non-occupational exposure. A conventional figure of 70 years has been adopted.

Appendix II—Relative Biological Efficiency

The relative biological efficiency of any given radiation has been defined by comparison with the gamma radiation from radium filtered by 0.5 mm of platinum. It has been expressed numerically as the inverse of the ratio of the dose of the two radiations (in ergs per gram of tissue) required to produce the same biological effect under the same conditions. It has been assumed, for purposes of calculation, that the relative biological efficiency of a given radiation is the same for all effects mentioned in the Introduction, with the single exception of gene mutations.

The following values have been adopted:

Radiation	*Relative Biological Efficiency*
Gamma rays from radium (filtered by 0.5 mm Pt) X-rays of energy 0.1 to 3.0 Mev Beta rays	1.0
Protons	10
Fast neutrons of energy not greater than 20 Mev	10
Alpha rays	20

The effective figure for slow neutrons should be derived in any given case from an evaluation of the separate contributions to the biological effect by protons arising from the disintegration of the nitrogen nuclei and by gamma rays arising from the capture of neutrons by hydrogen nuclei. —END

Behavior of Carrier-free Tracers—I *(Continued from page 59)*

thorium B and thorium X in potassium and rubidium sulfates varied from 15 to 30, some values even lying outside this range.

(2) The presence of a small amount of bismuth ion suppressed the incorporation of the tracer; for example, addition of 1 mg of bismuth reduced the value of D at 20° C for thorium X in potassium sulfate crystallized from 0.05 *f* sulfuric acid solution from ~25 to ~2.

(3) Radioautographs of the active crystals showed (*1*) that the tracers were regularly but discontinuously incorporated.

Hahn (*1*), who discusses coprecipitation by anomalous-mixed-crystal formation and "internal adsorption" in greater detail than is discussed here, points out that it is not always possible to distinguish between these carrying processes. He also mentions coprecipitation by occlusion of radiocolloids and active mother liquor. —END

NEW TYPE COUNTING-RATE RECORDER*

A standard scaler and recording millivoltmeter were modified to form an instrument that plots points directly proportional to arithmetic mean of count during fixed intervals

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IN MANY APPLICATIONS of G-M and scintillation counting, a suitable recording counting-rate meter can be very useful. In certain *in vivo* biological tracer experiments, for instance, the counting rate from one or more counters must be followed closely for hours, resulting in a quantity of data which requires much routine labor to record, compute, and plot by hand.

Most of the counting-rate meters used in the past have been such that the influence of any given count on the indicating meter decreases with time at an exponential rate after the occurrence of the count (*1*, *2*). An instrument of this type has the disadvantage that when a large change in the counting rate occurs, a relatively long time is necessary for it to reach equilibrium and thus indicate the new counting rate correctly. A special statistical theory is necessary to evaluate the statistical errors and transient response of such an instrument (*3*).

The recording counting-rate meter described here has several advantages. First, the recorder plots a series of points, each point representing the arithmetic mean of the counting rate during a given fixed interval of time. The regular statistical theory is then valid. In addition, the instrument uses a chart which has straight rectangular coordinates. The counting rate is plotted as a linear function of the input counting rate. Instrument error, exclusive of the fluctuations caused by random variations in counting rate, is less than 2% of full scale. Three or more curves from different G-M tubes or scintillation counters can be plotted simultaneously on one chart if desired.

Principle of Operation

Nearly all G-M and scintillation counting at this time is done with electronic scaling circuits combined with mechanical registers. Scaling circuits were used in this instrument since they make it possible to design a counting-rate circuit that is simple and highly accurate.

The instrument consists of a standard type of G-M scaler, a Leeds and Northrup Speedomax recorder, and the relay circuit shown in Fig. 1. The principle of operation is as follows: The pulses from the G-M tube or a scintillation counter go to the input of the scaling circuit in the usual manner. They are scaled down and then, instead of operating the mechanical register in the scaler, operate the high-speed relay shown in Fig. 1. The relay is a single-pole double-throw type with a small capacitor C_1 connected between the relay armature and ground. With the relay

* This work was performed under the auspices of the Atomic Energy Commission.

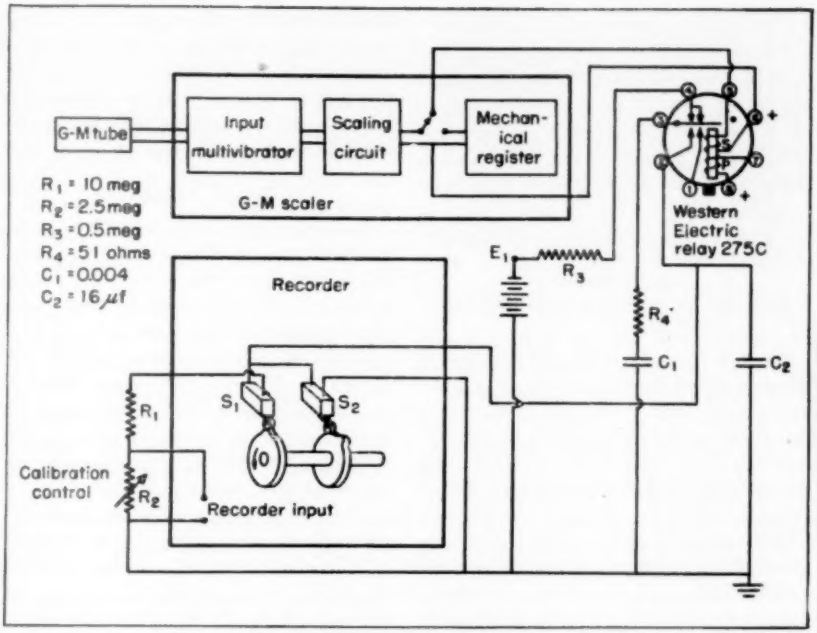

FIG. 1. Wiring diagram of counting-rate recorder

armature at its rest position, C_1 is fully charged by the fixed voltage E_1 connected to relay contact *4*. With each register pulse, the armature of the relay moves momentarily to relay contact *2*, and C_1 discharges into the large tank capacitor C_2. This causes the voltage across C_2 to increase by a constant increment with each relay click.

After a fixed interval of time, the switch S_1 closes and connects C_2, through a voltage divider, to the input terminals of the recorder. Switch S_1 is operated by a cam which is added to the recorder. After the voltage across the capacitor is plotted by the recorder, the cam moves to the next position and C_2 is completely discharged by S_2. Then the cam rotates further, S_2 is opened, and C_2 begins to integrate the relay clicks as before. This cycle of operations is repeated continuously, one point being plotted on the chart for each cycle. The position of each point is directly proportional to the number of relay clicks which have occurred and therefore to the counting rate during each interval of time.

The range of this instrument is changed simply by switching the scaling factor of the scaler. A scale-of-64 is used for a full-scale indication of 40,000 cpm, 32 for 20,000 cpm, etc., down to a scale-of-2 for 1,250 cpm. Still lower ranges can be had by retaining the scale-of-2 and switching in larger-value capacitors for C_1.

An example of the record obtained by this recorder from a scintillation counter is shown in Fig. 2. In this record, a point was plotted every 48 sec, so each point represents the average counting rate over a 48-sec period. Full scale is 40,000 cpm. The sample was first placed close to the counter for about an hour after which it was suddenly moved

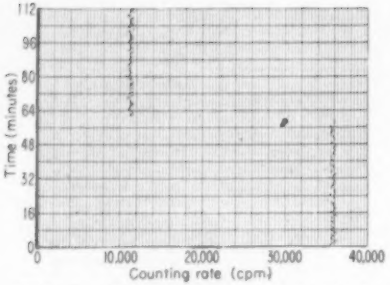

FIG. 2. Example of record obtained

further away. It was moved several seconds after the beginning of a time period. It will be noted that, when the counting rate changed, a point was plotted that is located between the former and latter counting rates. (This point is indicated by arrow in Fig. 2.) The reason, of course, is that the instrument always plots the average counting rate during each 48-sec interval without regard to whether there are any variations within that period.

To follow more rapid variations in counting rate, the printing time interval must be made shorter. Of course, the statistical variation of the points will then be greater. The time interval can be changed to 24, 12, or 6 sec by substituting different cams with the proper number of teeth. Other intervals can be obtained by making changes in the original recorder mechanism.

Details of Design

In the system built here, a Nuclear Instrument and Chemical Corp. model 163 scaler and a Speedomax model G, 6-point, 0–10 mv recorder are used. The cam and cam-operated switches which were added to the recorder are shown in Fig. 3. The cam position must, of course, be adjusted so that its operation is synchronized with the switching of the recorder between its six input terminals.

It will be noted that the accuracy and stability of this circuit is dependent primarily upon the value of the resistors R_1 and R_2, the capacitors C_1 and C_2, and the voltage E_1, assuming that the scaler and recorder are operating normally. The accuracy is not particularly dependent on the characteristics of any vacuum tubes or of the relay, as long as the register pulses do not arrive at

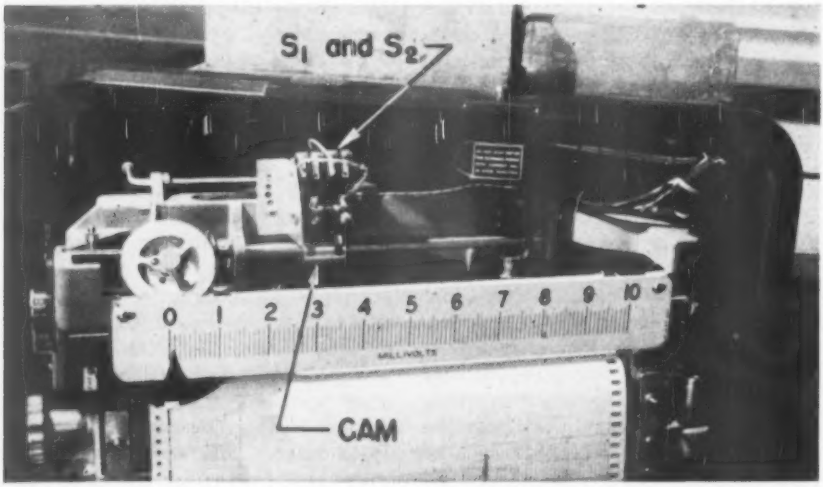

FIG. 3. Modified recording millivoltmeter showing installation of switches and cam

such a high rate that the relay is not able to make contact for each pulse. The length of time the relay makes contact is not important since C_1 discharges completely into C_2 in a very short time. According to the manufacturer, the relay has been designed to follow a uniform pulse rate of 100 pulses per second, much higher than necessary.

It is a characteristic of mercury relays, such as the Western Electric 275C, that the armature makes contact momentarily with the new contact before it breaks with the previous one. This means that current from the voltage source E_1 can flow for a short time directly into C_2. Fortunately, the length of time during which current can flow in this way is small. The resistor R_3 is used to limit the magnitude of this current. This resistor should not be made too large or it will prevent C_1 from charging fully when the relay armature is in the rest position. A trial of several relays showed that the charge transferred to C_2 by the momentary short circuit was constant for each relay, although the amount varied with different relays. The charge transferred by the momentary short circuit was in the order of only a few percent.

The net effect of this momentary short circuit is that C_2 gets a small extra charge with each relay click. As long as each short circuit lasts the same length of time, the over-all accuracy of the instrument is not impaired. This seems to be the case in actual practice. The mercury relay was used in spite of this characteristic because of its high speed and quiet operation.

Linearity

There are several things which must be considered in the design of this instrument for it to be linear. First, the value of C_1 should be very small in comparison with C_2 so that the voltage built up across C_2 in each time period is only a small fraction of the supply voltage E_1. Otherwise the charging curve for C_2 and the indication of counting rate will begin to be appreciably nonlinear. By using the circuit values given in Fig. 1, the deviation from linearity is less than 0.8%.

The linearity will also be affected if R_3 or R_4 are too large, because there will not be enough time for C_1 to charge and discharge completely during the cycle of operation of the relay. If the values given in Fig. 1 are used, however, the nonlinearity introduced by this effect is completely negligible.

Calibration of the instrument is accomplished by adjustment of R_3. Once this is set for one sensitivity range, it will be just as accurately set for all the ranges as long as only the scaling factor is changed. If the value of C_1 is changed to provide more sensitive ranges, means should be provided to switch in different values of R_2 to compensate for slight inaccuracies in the values of C_1.

* * *

The author wishes to acknowledge the interest and encouragement of Dr. C. A. Tobias.

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"Studies have indicated that only 15 to 20 per cent of those trained in scientific, engineering, and technical fields are actively engaged in research and development work."

—Eric A. Walker, Executive Secretary, Research and Development Board, speaking at the AAAS Symposium on the "Effects of Government Support on Scientific Research," Cleveland, Ohio, Dec. 30, 1950

BOOKS

Clinical Use of Radioactive Isotopes, by Bertram V. A. Low-Beer, Charles C Thomas, Springfield, Ill., 1950, xv + 414 pages, $9.50. *Reviewed by John P. Storaasli, M.D., University Hospitals of Cleveland, Ohio.*

The author has divided his text into two parts.

Part One deals with a review of the physical aspects of the atom, including its mass, energy, and charge relationships. The chapter on radioactivity, although somewhat brief, is well written and will give those readers who are new in this field adequate knowledge of the most important properties of radioactive isotopes. In this section is included a description of the various types of nuclear radiations, the production of artificial radioactivity, and the laws of radioactive decay.

The section on detection and measurement of radioactivity gives a review of autoradiographic techniques for tissues. The various techniques are outlined in respect to type of particle and proper photographic emulsion to be used. Other methods of detection and measurement are also included. The author discusses in some detail, the statistical analysis of counting data in relation to background, total number of counts, counting rate, and circuit used.

The last chapter in Part One deals with health physics. It gives data on storage and disposal of radioactive materials. Tables of tolerance amounts of isotopes commonly used clinically are presented. Part One is well illustrated and has a good bibliography.

Part Two deals with the use of radioactive isotopes in clinical investigation. The isotopes most commonly used in medicine are discussed in detail and the reader can find some of the most recent and salient uses of such elements as radiophosphorus, sodium, iron, iodine strontium and calcium. The production, physical characteristics, general distribution and excretion patterns of each of these radioelements are concisely outlined.

The uses of radiophosphorus in the study of phospholipid metabolism, in studies of circulation, and in determining blood volume are covered. Its use as a diagnostic aid in breast carcinoma and brain tumors is also discussed. Radiosodium as a diagnostic adjunct in studies of peripheral vascular disease, extravascular space and of shock states is presented, together with an unbiased condensed summary of the available literature on the use of radioiodine in thyroid physiology and as a diagnostic aid in hyperthyroidism. Brief accounts of the uses of radioisotopes of arsenic, mercury and radiocolloids of gold, zinc and manganese are given.

Chapter III of Part Two is a survey of the literature and the author's own experiences in using radioisotopes in the treatment of disease. Radiophosphorus in the treatment of the leukemias, polycythemias and lymphomas is dealt with, as is its use as a source of external radiation.

No text of this nature would be complete without space devoted to the use of radioiodine in the treatment of hyperthyroidism and thyroid cancer. Again, the author surveys the literature and gives his own experiences in this field with an over-all picture of the problem involved.

A few pages are devoted to the use of radiocobalt as a source of external and interstitial radiation.

The last fifty pages of the text are devoted to a comprehensive bibliography of over three hundred references. Nine appendices are presented dealing with tables of isotopes, gamma-ray shielding, and dosage computations.

Introductory Nuclear Physics, by David Halliday, John Wiley & Sons, Inc, New York, 1950, ix + 558 pages, $6.50. *Reviewed by Arthur Roberts, Department of Physics, University of Rochester.*

In 1931 there appeared, most opportunely, a new edition of Rutherford, Chadwick, and Ellis's "Radiations from Radioactive Substances." It came out just before the explosive developments in nuclear physics of 1932–1934, which included the first disintegrations produced by artificially accelerated particles, the discoveries of the neutron, deuteron and positron, and the discovery of artificially produced radioactivity. Thus, for the new students attracted to the incandescence of nuclear research, it summarized admirably what was known in the field at that time, the conclusion of the classical era of nuclear physics.

Since then it has become impossible for any one book to cover the whole field of nuclear physics. Bethe's famous articles in the *Reviews of Modern Physics* in 1936 and 1937 covered the field of nuclear theory, and remain, in fact, the best available single work in that field. It is, however, a little surprising that since Rutherford, Chadwick, and Ellis, no one has produced an adequate survey of nuclear physics at the descriptive, introductory level. It is just such a survey that has now been provided by Halliday's new book.

The book is intended as a text for first-year graduate students or seniors with some acquaintance with atomic physics. It treats primarily of the experimental side of nuclear physics, and only glances at the highlights of the theory. To cover the field, the treatments of most subjects necessarily had to be brief. They are concise, accurate, and illuminating. The treatment of experimental detail is thorough, even to the point of giving circuits for scalers, blocking oscillators, and similar useful gadgets. All the important experimental techniques are here, clearly and accurately described, and beautifully illustrated in figures. There are many problems and exercises.

The book is not intended, nor will it be useful, for an advanced graduate course in nuclear physics. No up-to-date text to suit this purpose exists.

One secondary purpose of the book is as a first source book in experimental methods. It can be particularly recommended for this purpose to advanced theoretical students, whose heads are often lost in the clouds of field theory. The book should also be very useful for the specialist in fields other than nuclear physics, who is interested in nuclear technology. Provided he has already mastered the fundamentals of atomic physics, he will find here a good account of nuclear techniques as they are practiced in the laboratory.

There are a few minor objections to be made. The almost complete lack of historical development may puzzle the thoughtful student, who will not understand how a particular subject has reached its present state of development. Use of only the most recent references in many subjects often fails to give perspective or to allocate properly the credit for important work. This fault is not trivial in a textbook which will be widely used; it offends an important motivation of scientists, and is not readily remedied. The word *nuclide*, meaning *nucleus*, appears to me both ugly and unnecessary; I am sorry to see it perpetuated here. None of these objections is serious in comparison with the many virtues of the book.

BOOKS RECEIVED

Toxicology of Uranium, Survey and Collected Papers, (Div. IV-23 of the National Nuclear Energy Series) edited by Albert Tannenbaum, McGraw-Hill

(*Continued on page 91*)

CROSS SECTIONS

Continuously Sensitive Diffusion Cloud Chambers

After over a decade of inactivity, interest has been revived in diffusion cloud chambers which allow continuous observation of the tracks of an ionizing particle. The first cloud chamber operating in this manner was reported by A. Langsdorf (*1*) in 1939. Because expansion chambers gave better results, however, they have been used almost exclusively in the intervening years.

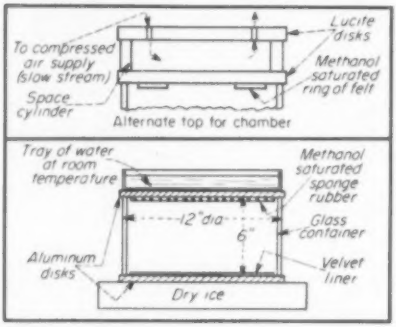

FIG. 1. Diagram of simple diffusion cloud chamber. Alternate top is used for photographing tracks through top of chamber

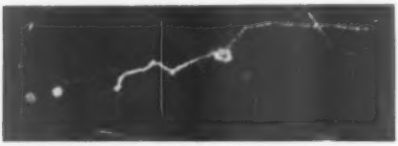

FIG. 2. Diffusion cloud chamber photograph of track of heavily ionizing particle

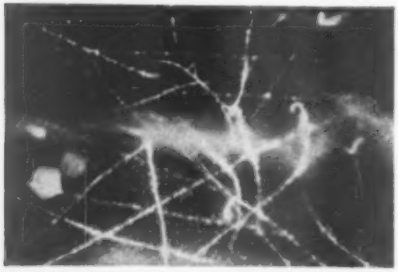

FIG. 3. Diffusion cloud chamber photograph of tracks produced by radiation from gamma-ray source

Recently, E. W. Cowan (*2*) of the California Institute of Technology devised a simple diffusion chamber. Almost simultaneously, investigations were begun at the University of California Radiation Laboratory, Yale University, Massachusetts Institute of Technology, Ohio State University, Brookhaven National Laboratory and elsewhere.

A diagram of the basic cloud chamber is given in Fig. 1. The temperature gradient between the room-temperature top and dry-ice-cooled bottom creates partial pressures which cause the methanol vaporizing from the sponge rubber to diffuse toward the bottom of the chamber. At some point between the two surfaces the vapor is cooled sufficiently to produce supersaturation, a required condition if the vapor is to condense on particle tracks. A light shined through the cylinder illuminates the tracks.

Metal collars may be placed around the bottom of the chamber to adjust the temperature gradient. Best operation occurs with average gradients between 3° and 7° C/cm.

Photographs of particle tracks obtained with this type of chamber are shown in Figs. 2 and 3. Scientists at Brookhaven National Laboratory used the alternate top for the chamber shown in Fig. 1 in making these photographs.

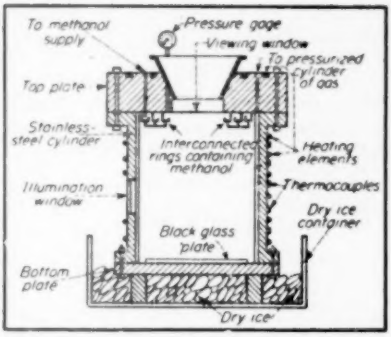

FIG. 4. Diagram of high-pressure diffusion cloud chamber

At Brookhaven, Drs. D. H. Miller, E. C. Fowler, and R. P. Shutt are extending their investigations to high-pressure diffusion cloud chambers which will be used to study nuclear interactions in gasses. Particle tracks have been obtained in hydrogen at pressures up to 15 atmospheres (*3*). The high-pressure chamber is made of stainless steel as shown in Figs. 4 and 5. Electric heating elements and thermocouples are used to control the temperature gradient. Thick glass windows are provided for viewing and illumination.

It is expected that a phototube can be used to trigger a camera when the chamber contains a multitude of tracks. This would eliminate the need for continuous observation in certain studies.

Advantages

Diffusion cloud chambers are not expected to replace expansion types, but they offer advantages in certain applications. Their almost continuous sensitivity makes them valuable in monitoring accelerators, etc., where pulses of radiation occur more frequently than an expansion chamber can be recycled. Because of their simplicity, they are ideal for instruction purposes.

Disadvantages

There are several disadvantages inherent in these chambers. First, the sensitive layer has a maximum depth

FIG. 5. High-pressure diffusion cloud chamber. Stainless-steel tank is wrapped with insulation

of 4–5 in. that is limited by temperature gradient. High temperatures cause fog and instability. This limits observable track length in cosmic-ray studies. Sec-

ABOUT THE COVER

TRACKS of secondary electrons produced by gamma rays from a Co^{60} source held near a diffusion cloud chamber are visible in the cover picture. They were clear in the original print, but were retouched to compensate for loss in engraving. Heating element in top pan was used to keep water at room temperature. Photo from Brookhaven National Laboratory.

ond, metal plates cannot be inserted as absorbers or for production of secondaries because they interfere with diffusion. This may possibly be overcome by heating the plates with electric elements. Third, background cannot be too large. The chamber cannot be swept out with an electric field without disrupting operation. Fourth, after heavy ionization, 5–10 sec are required for revaporization. During that time the chamber is insensitive.

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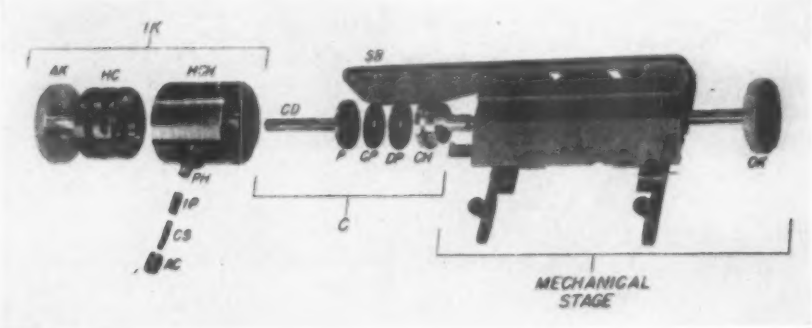


Microscope Stage Accessory for Track Counting*

By Richard C. Roth

Brookhaven National Laboratory
Upton, New York

One of the techniques used in neutron monitoring involves the counting of proton recoil tracks in nuclear emulsions. The Health Physics Division of the Brookhaven National Laboratory employs a Bausch and Lomb research type microscope, Model DDE, with a 10X eyepiece and a 1.8 mm (98X) oil objective for counting these tracks in Eastman N. T. A. films. The maximum permissible dose has been expressed for this film as a certain maximum allowable number of tracks per field. Twenty-five fields of 0.15 mm each are routinely scanned.

An accessory has been built for the microscope which automatically and accurately moves the mechanical stage exactly one field 25 consecutive times. It may be adapted for any field size and for any predetermined number of fields, and may be of use in other endeavors employing a microscope.

An indexed knob is substituted for one of the regular knobs of the mechanical stage. It does not interfere with ordinary use of the microscope, and the only modification required of existing parts is the drilling and tapping of four small holes in the stage.

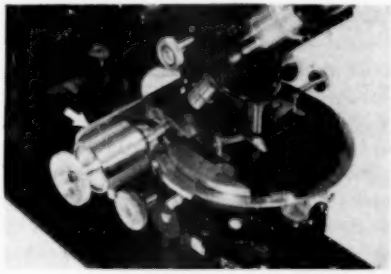
The modified mechanical stage and an exploded view of the accessory (indexed knob *IK* and the clutch *C*) are shown in the illustration above. One of the original knobs, *OK*, may still be

* Research carried out under auspices of the U. S. Atomic Energy Commission.

used normally. When it is turned, the friction between the index pin *IP* and the indents of the helical cam, *HC*, is great enough to cause the clutch to slip and keep *IK* from being actuated. When the adjusting knob *AK* is turned, the rotary motion is imparted through a square clutch drive *CD*, within it, to a brass plate *P*. This plate frictionally engages a rubber clutch plate, *CP*, which in turn transfers the motion to a brass driven plate, *DP*. The ends of four small compression springs butt into four circular recesses in *DP*. These springs are loosely mounted coaxially over screws threaded into the clutch housing, *CH*. This housing is the actual substitute for the other original knob of the stage. Held in place on the shaft of the stage by a set screw, *CH* turns the lead screw in the desired controlled motion.

This controlled or "stopped" motion is provided by a cam and index pin. This brass cam, *HC*, with a $1\frac{5}{16}$-in. outside diameter, has a helical slot $\frac{3}{32}$-in. deep milled for a total of 500 degrees along its lateral surface, with a lead angle of 5° 26′ 24″. On the floor of this slot are 25 V-shaped indents $\frac{7}{64}$-in. wide, spaced 20° 28′ 45″ apart. These indents are engaged by the spring-loaded index pin, *IP*, which acts as a stop. Difficulty of turning knob *AK* is adjusted by means of the compression springs, *CS*, and the adjusting cap, *AC*. The helical cam fits inside a stainless steel housing *HCH*, and the pin assembly mounts in *PH*, the pin housing.

The whole accessory is coupled to the stage with the support bracket, *SB*. Six screws threaded into a flat milled on the back of *HCH* hold the accessory and bracket together; *SB* itself is held onto the stage by four small screws.



The illustration above shows the assembly mounted in place. The whole affair is solid but light and is easily removable.

TECHNICAL BRIEFS

• **Pump** for liquid metals uses pressure created by magnetic field on corrosive molten metal through which electric current is being passed. In effect, the metal pumps itself.

• **Fluorimetric** method for determining uranium in low-grade ores has been developed by F. S. Grimaldi and H. Levine of the U. S. Geological Survey. The procedure employs batch extraction of uranium nitrate by ethyl acetate, using aluminum nitrate as the salting agent, prior to visual estimation. Working range is 0.001 to 0.04% U.

• **Double-nuclei white blood** cells appear "to be an unusually promising means of identifying potentially harmful operations before gross overexposure can occur," according to Drs. M. Ingram and S. W. Barnes of the University of Rochester. They have found that exposures below tolerance cause single-cored cells to change.

• **Dosimeters** developed by the Navy utilize color change in alkali halide crystals and in silver-activated phosphate glass to indicate exposure to gamma rays. Crystals turn blue; glass turns orange. The depth of color is proportional to the dose of radition received.

NUCLEONIC EVENTS

AEC O.K.'s MONSANTO AND DOW-DETROIT EDISON PROPOSALS TO STUDY FEASIBILITY OF DUAL-PURPOSE REACTORS

In a carefully worded statement, the Atomic Energy Commission has announced its approval of proposals by two industrial groups to study the feasibility of dual-purpose nuclear reactors. At the same time, the AEC has recommended a guide for other industrial concerns to follow in making similar proposals. The proposals approved were one presented by C. A. Thomas, executive vice president of the Monsanto Chemical Company, and one jointly presented by J. W. Parker of the Detroit Edison Company and M. E. Putnam of the Dow Chemical Company. Both of these groups are interested in studying the feasibility of developing and operating nuclear reactors for the production of plutonium and with the ultimate goal of producing electric power.

It is reported that the proposals made by the two groups actually consist of three steps:

1. Feasibility study
2. Design
3. Construction and operation

However, the AEC's announcement states that the "Commission has also directed that no commitment be made at this time to continue beyond the study phase."

As far as further proposals from industrial concerns or groups, "the Commission has specified that those submitting such proposals should put forth a well-defined set of objectives for their proposed study, should be financially responsible and equipped with a staff of adequate qualifications and size, and should undertake to submit a report including detailed findings and recommendations at the conclusion of the study. These reports would be disseminated at the discretion of the Commission to the extent consistent with security."

The AEC expects to permit "properly qualified and cleared persons" (probably not exceeding 10–15 individuals from each group) to have access to existing information and to take up "a reasonable share of the time of Commission personnel."

In considering these proposals, the Commission was guided by the need that they benefit the atomic energy program in from one to three years. Also, it was important that the concerns involved would undertake their studies on their own initiative, using their own resources.

The Commission expressed the hope that these proposals "may ultimately lead to arrangements where, because initiative is with industry, additional incentives for rapid and aggressive technical and business development will exist."

Thomas Proposal

Dr. Thomas' proposal was first announced in June and published in NUCLEONICS in August, 1950 (p. 73). At that time, he said that the contemplated power plant would be built by industry with its own funds.

He also said, "The uranium, the necessary source of power, would be

loaned or leased by the government to the industrial plant, which would convert a portion of it into plutonium and power. The power would belong to the industry that put up the money, but the plutonium and the isotopes and all the fissionable by-products would at all times belong to the government."

Dr. Thomas contemplates that the government would send the canned uranium to the industrial reactor for fuel. After a period of reactor operation, the cans would be removed by the government and processed at its own plants for recovery of plutonium. Industry would receive a fee for this plutonium which would go toward offsetting its costs.

AEC SIGNS CONTRACTS FOR PADUCAH PLANT

The Atomic Energy Commission has signed contracts with four concerns for architect-engineer and construction work on the AEC's new uranium-235 production facility at Paducah, Kentucky.

F. H. McGraw and Co. of Hartford, Conn., will be the principal construction contractor. The McGraw Company will build all structures at the site and install production equipment. Giffels and Vallet, Inc., of Detroit will handle preliminary engineering and design of the process plant. Sargent and Lundy, Chicago, will make power studies and design power facilities. Smith, Hinchman and Grylls, Inc., Detroit, will do the design and engineering for the sanitary and fire water systems, sewer system, treatment plant and steam plant.

TRUMAN RECOMMENDS '52 BUDGET OF $1,277-MILLION FOR AEC

In the budget message he sent to Congress last month, President Truman recommended estimated expenditures of $1,277-million for the nation's atomic-energy program in the fiscal year 1952. He estimated expenditures for fiscal year 1951 as $818-million, as against actual expenditures of $550-million for 1950.

"At the same time that we are actively pursuing industrial and other peacetime applications of atomic energy," the President explained, "present world developments demand intensification of the national security aspects."

REACTOR TECHNOLOGY SCHOOL ACCEPTING APPLICATIONS

The Oak Ridge School of Reactor Technology is currently receiving applications for enrollment in the 1951–52 session, beginning September 10, 1951. This School was established at the Oak Ridge National Laboratory in March of 1950 (NU, Jan. '50, p. 95).

The school will enroll students of outstanding qualifications who hold, or will receive by September 1951, bachelor's or master's degrees in chemistry, engineering, metallurgy or physics. A limited number of recent college graduates will be accepted under Category A in the status of student-employees of the Oak Ridge National Laboratory, and will be paid a monthly stipend for a twelve-month period, beginning September 1951.

Provision is also made for trainees sponsored by government agencies and industrial organizations connected with or interested in the AEC reactor development program, Category B. Applications for enrollment under this category must be made by the firms or agencies employing the applicants. Such students remain on the payrolls of their home organizations.

All enrollments are contingent upon a personnel security investigation.

Further information and application forms may be obtained by writing to the Director, Oak Ridge School of Reactor Technology, Post Office Box P, Oak Ridge, Tennessee. Category for which application is requested must be specified. Academic and industrial ap-

plications must be filed with the director of the school by March 1 and April 1, 1951, respectively.

SHUGG RESIGNS AEC DEPUTY MANAGERSHIP

Carleton Shugg on January 31 resigned from his post as deputy general manager of the Atomic Energy Commission. Long associated with shipbuilding before coming to the AEC, Mr. Shugg will become assistant general manager of the Groton, Conn., plant of the Electric Boat Company which is constructing the hull for an atomic-powered submarine.

A graduate of the Naval Academy at Annapolis, he worked for the Sprague Electric Company, the Cramp Shipbuilding Company, the Todd Shipbuilding Corporation and the Hanford, Wash., works of the AEC before taking the AEC deputy general managership in Washington.

Last August, Congress appropriated $350-million for shipbuilding that included authorization for the nation's first atomic-powered submarine. Two separate projects to develop a dry-land model of a nuclear marine engine are currently being worked on for the AEC by the Westinghouse Corp., cooperating with the Argonne National Laboratory, at Arco, Idaho, and by the General Electric Company at Schenectady, New York.

IRE SCHEDULES PAPERS ON NUCLEAR SCIENCE, REACTORS

The 1951 National Convention of the Institute of Radio Engineers to be held at the Waldorf-Astoria Hotel in New York, March 19–22, has scheduled a session on nuclear science and a symposium on reactors as part of its March 22 program.

The nuclear science session, to be held during the morning, includes the following papers: *A Delayed Coincidence Scintillation Spectrometer*, by F. K. McGowan; *Timing Unit and Pulse Deflector Generator for 145-inch Synchrocyclotron*, by E. M. Williams, C. H. Grace, and L. W. Johnson; *Design and Construction of a Billion-volt Linear Electron Accelerator*, by M. Chodorow, E. L. Ginzton, J. Jasberg, R. Kyhl, R. Neal, and P. Pearson; *Precise Measurement and Regulation of Magnetic Fields with RF Techniques Using Nuclear Resonance*, by H. A. Thomas; and *A High Precision Magnetic Field Measuring Instrument*, by E. C. Levinthal.

Participants and their subjects for the symposium on nuclear reactors, to take place during the afternoon session, include: A. M. Weinberg, *Nuclear Energy Development and Military Technology;* D. Hughes, *The Reactor as a Research Tool;* F. P. Cowan, *Background Radiation Monitoring for Control of an Air-cooled Pile;* and J. E. Binns, *Instrumentation in the Brookhaven Nuclear Reactor.*

AEC OFFERS FELLOWSHIPS IN INDUSTRIAL MEDICINE

Continuing a program begun last September, the AEC will offer four fellowships in industrial medicine during the 1951–52 academic year. The fellowships provide advanced training to men and women medical doctors in the field of industrial medicine, in relation to the atomic energy industry.

Awards of $3,600 plus tuition and fees are made for one year's academic training at any approved institution. Upon completion of this training, fellows will be eligible to apply for a second year of in-plant training at one of the major AEC installations. Stipend for the in-plant year is $5000.

Fellowships are open to U. S. citizens who hold an M.D. degree from an approved medical school and who have had at least one year of internship. Applications should be submitted by March 1, 1951 to AEC Fellowships in Industrial Medicine, Atomic Energy

Project, School of Medicine and Dentistry, University of Rochester, Rochester, N. Y.

CANADA TO BUILD NEW, MORE POWERFUL REACTOR

Last month Trade Minister C. D. Howe announced that Canada is going to build a new $30,000,000 nuclear reactor at Chalk River, Ont. The new development, he said, should put Chalk River eventually on a paying basis by sales of increased supplies of plutonium and radioisotopes.

The new reactor was described as "many times more powerful" than Chalk River's present NRX reactor, which is a complex cylinder 34 feet high, capable of delivering the highest neutron flux known for existing reactors. Like NRX, the new reactor will be heavy-water moderated.

This will be Chalk River's third reactor. The first was the ZEEP, which recently came under declassification along with other low-power reactors and is still in use as a partner to the NRX pile.

Breeding and power are among problems to be attacked in connection with the fundamental research planned with the reactor. It will be both a commercial and a research project, in contrast to the experimental character of the present reactor.

Preparations for the new development will start immediately at Chalk River, but actual construction will not begin until late summer. Chalk River already is stockpiling plutonium and selling radioisotopes, but production of both will be stepped up sharply when the new reactor is finished, probably sometime in 1952.

IN BRIEF

➤***The TVA** will build a steam electric power plant on the Kentucky side of the Ohio River fifteen miles northwest of Paducah, site of the new atomic energy installation which it will serve. The Tennessee Valley Authority plant will be the second power supply planned for the site, the first being the $65,000,000 steam-generating facility to be built by Electric Energy, Inc., a five-company concern organized in December (NU, Jan. '51, p. 84) and recently authorized by the Securities Exchange Commission.*

➤**Closer cooperation** with foreign nations in atomic matters is being studied by the AEC. According to AEC's chairman Gordon Dean, Congress will receive the Commission's recommendations in the matter shortly. Although the individual nations cannot be named in any resulting legislation, the countries AEC has in mind are Canada and the United Kingdom. The cooperation would include a trade of men and materials as well as technical secrets.

➤***Applications for employment** with either the du Pont company or the AEC can now be filed each Friday in Barnwell, S. C., in connection with the Commission's new $260-million production facility to be built there. Applications are also accepted each Wednesday in Aiken at the U. S. Employment Service office.*

➤**Graduate fellowships** in physics, chemistry, metallurgy, ceramics, mechanics, and electrical engineering, to begin in September, are being offered by the Armour Research Foundation

REPRINTS available of "Lab Equipment Guide"

A limited number of reprints of the special section published in the November NUCLEONICS devoted to a "Guide for Selection of Equipment for Radioactivity Laboratories" and "Buyers' Guide" are available and can be obtained by sending $1 for each copy desired to: Editorial Department, NUCLEONICS, 330 West 42d Street, New York 18, New York.

of Illinois Institute of Technology, Chicago, Ill. Applications received prior to March 15 will be given first consideration.

➤***The Phoenix Project,*** *devoted to research into the peaceful use of nuclear energy, last month received a grant of $100,000 from the Nash-Kelvinator Corporation to be applied toward basic research on the preservation of food and improvement of metal surfaces and coatings. The Project is administered by the University of Michigan.*

➤**Visits of lecturer-physicists** to the University of Rochester have been made possible through funds provided by a group of Rochester, New York, industries. Wolfgang K. H. Panofsky, theoretical physicist at the University of California's Radiation Laboratory, was the first visiting lecturer brought to Rochester under the arrangement.

➤***BEPO,*** *the larger of England's two nuclear reactors at Harwell, will soon have an output of 10,000 kw instead of the present 6,000 kw. This, according to Sir John Cockroft, Harwell's director, is the expected result of recent promising metallurgical developments.*

➤**Paducah plant headquarters** of the AEC were formally opened on January 15 at the site of the old Kentucky ordnance works where the Commission's new gaseous diffusion plant will be built. The Carbide and Chemical Division of Union Carbide and Carbon Corp. has opened an employment office in Paducah in connection with its contract to operate the plant.

➤***Drawings and estimates*** *of a prototype atomic power unit for large merchant ships and warships are reportedly being made up by an engineering firm under contract to the British government.*

NUCLEAR NEWSMAKERS

William E. Keller has joined the staff of the chemistry and metallurgy division the Los Alamos Scientific Laboratory as a research chemist. He was formerly with the Ohio State University Research Foundation.

Gail D. Adams is joining the staff of the University of California School of Medicine as associate physicist on biological and medical research.

Hideki Yukawa, Japanese Nobel Prize winner for his theory on the meson, has decided to remain in this country and accepted appointment as full professor of physics at Columbia University. He has been at the University since 1949 as a visiting professor.

Garth W. Edwards has been appointed chief of the administrative division of AEC's new production plant at Aiken, S. C. He was formerly with the AEC's procurement division in Washington.

C. W. Tittle is now associated with the Gulf Research and Development Company in Pittsburgh. Until last month he was on the faculty of North Texas State College in the department of physics.

Andrew Kalitinsky, formerly with the Fairchild NEPA Project at Oak Ridge, has become technical assistant to the vice-president in charge of engineering of the M. W. Kellogg Co.

NECROLOGY

Yoshio Nishina (January 10, 1951). Dr. Nishina was pre-war head of the Japanese Physico-Chemical Research Institute, served as president of the Science Research Institute in Tokyo, and as vice-president of the Science Council of Japan. He is credited with producing the first uranium-237 in Japan and was instrumental in developing the Klein-Nishina formula on Compton effect.

MEETINGS

American Society of Mechanical Engineers, Nuclear Energy Division of Metropolitan Section—New York, N. Y., Feb. 20

American Mathematical Society, 466th Meeting—New York, N. Y., Feb. 24

Pittsburgh Conference on Analytical Chemistry and Applied Spectroscopy—William Penn Hotel, Pittsburgh, Pa., Mar. 5–7

American Physical Society—Pittsburgh, Pa., Mar. 8–10

National Association of Corrosion Engineers, Annual Meeting—Hotel Statler, New York, Mar. 13–17

Institute of Radio Engineers, National Convention—Waldorf Astoria Hotel, New York, N. Y., March 19–22

American Chemical Society, 119th National Meeting—divided, Boston, Mass., Apr. 1–5; Cleveland, Ohio, Apr. 8–12

American Society of Mechanical Engineers, Spring Meeting—Hotel Biltmore, Atlanta, Ga., Apr. 2–5

The Electrochemical Society—Wardman Park Hotel, Washington, D. C., Apr. 8–11

Books Received *(Continued from page 81)*

Book Co., New York, 1951, xxvi + 333 pages, $3.

The Atom at Work, by Jacob Sacks, D. Van Nostrand Company, Inc., New York, 1951, xii + 327 pages, $4.

Applied Nuclear Physics (2d edition), by Ernest Pollard and William L. Davidson, John Wiley & Sons, Inc., New York, 1951, ix + 352 pages, $5.

Radiation Monitoring in Atomic Defense, by Dwight E. Gray and John H. Martens, D. Van Nostrand Company, Inc., New York, 1951, iv + 122 pages, $2. No technical background is necessary to understand or use this practical manual for radiological defense personnel. It gives a detailed and well illustrated account of the use of some standard radiation detectors and the interpretation of their results, and contains an appendix on standard calibration sources.

The Hell Bomb, by William L. Laurence, Alfred A. Knopf, New York, N. Y., 1951, 198 pages, $2.75. Written by the science reporter for *The New York Times* and twice winner of the Pulitzer Prize, this book presents in an orderly and non-technical fashion most of the facts on the hydrogen bomb that heretofore were not commonly available outside scientific circles. Some social and political questions relating to the bomb's use are discussed.

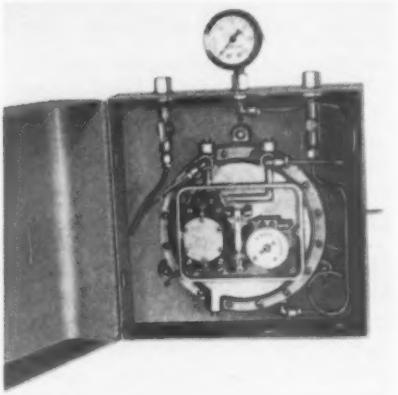
OTHER LITERATURE

Manual of Analytical Methods for the Determination of Uranium and Thorium in Their Ores, prepared by the technical staff of the Atomic Energy Commission's Analytical Laboratory at New Brunswick, N. J., under the direction of the Raw Material Operations Office.

This manual is intended principally for chemists and assayers with an understanding of chemical fundamentals. It was prepared for the main purpose of assisting and encouraging commercial laboratories, as part of their normal business, to take over gradually the function of making uranium analyses for prospectors and miners. This function is now performed largely by official laboratories of the AEC, U. S. Geological Survey and Bureau of Mines. *Available from Superintendent of Documents, U. S. Govt. Printing Office, Washington 25, D. C., $0.20.*

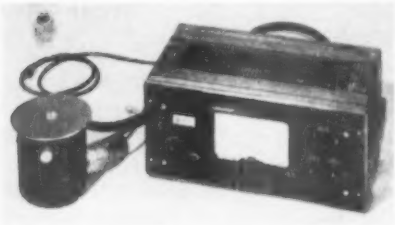
An International Bibliography on Atomic Energy, Vol. 1 (Political, Economic and Social Aspects), Supplement No. 1, 1950, 22-page pamphlet. The Atomic Energy Commission Group of the Department of Security Council Affairs, United Nations, supervised preparation of the supplement. *Available from the International Documents Service, Columbia University Press, New York 27, N. Y., $0.25.*

PRODUCTS and MATERIALS



AIR VELOCITY CONTROLLER

Johnson Service Co., 507 E. Michigan St., Milwaukee 2, Wis. Designed for control of air velocity through the doors of fume hoods, this proportional action velocity controller operates in the range of 50–250 ft/min. Accuracy is within ±10 ft/min. The difference in air pressures inside and outside the hood is used to actuate a pneumatic system which controls dampers or other controls in the ventilating system.



CAPACITOR ELECTROMETER

Loudon Instruments, Inc., 5644 Lake Park Ave., Chicago 37, Ill. Current, voltage, and rate-of-charge measurements can be made on low-current high-resistance circuits with the model 361–20 electrometer. Voltage ranges are 0–10, 0–100, and 0–1,000 mv. Multiple input resistors (10^9, 10^{10}, and 10^{11} ohms) with selective switching and grounding key are built into the head of the instrument. Sealed in a separate compartment inside the head is a dynamic capacitor used to convert the d-c input to an a-c signal which is then amplified, demodulated, and measured in a feedback circuit. The head can be located remotely from the amplifier. Provision is made for balancing contact potential and for switching input polarity. Terminals are provided for the connection of standard recorders.

FINGER CHAMBER

Landsverk Electrometer Co., Pippin Road, Cincinnati 31, Ohio. For protection of personnel working with X and gamma radiation, the model L-50 ionization chamber indicates hand exposure of 0–2 r. Calibration is said to be substantially independent of the energy of the radiation from 75 kv to 1.5 Mev. The hermatically sealed

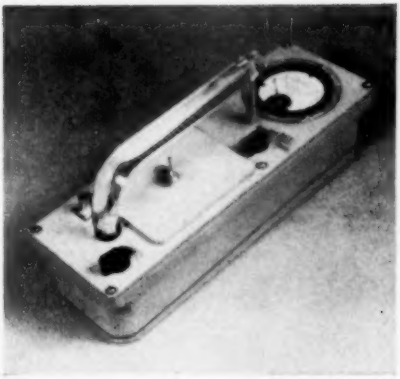
chamber is ½ in. long, 1 9/16 in. in diameter, and weighs ⅙ ounce. Finger rings are adjustable; chambers are also furnished with chains or clips. The chamber is charged and read on a battery-operated charger-reader, model L-60.

LINEAR AMPLIFIER

Nuclear Instrument and Chemical Corp., 223 W. Erie St., Chicago 10, Ill. Designed to adapt G-M scalers to proportional counting, the model 1061 linear amplifier provides sensitivity adjustment calibrated in millivolts. Maximum sensitivities of 1 or 10 mv can be obtained. Frequency response of the instrument is said to be flat from 10,000 cycles to 1.5 megacycles. A set of oscilloscope terminals are provided on the front panel. The scaler can be operated independently without disconnecting it from the amplifier.

MERCURY CLEANER

Bethlehem Apparatus Co., Front and Depot Sts., Hellertown, Pa. A motor-driven oxifier is used in conjunction with a gold adhesion filter to provide a purification system which will clean 5 pounds of mercury at a time. Base metal contaminations are oxidized by agitation in the oxifier for periods of 2 to 4 hours. A Bethlehem type G filter separates the oxides from the clean mercury.



ALPHA SURVEY METER

General Electric Co., Electronics Park, Syracuse, N. Y. The model 4SN10A2 portable alpha survey meter indicates 0–10,000 dpm on its semilogarithmic scale with an accuracy of ±10%. Calibrated for operation in the 4–5 Mev energy range, the instrument uses an air proportional counting chamber with a sensitive area of 6 × 20 cm. A thin window of less than 2 mg/cm² allows alpha particles to enter the chamber volume. Minimum relative geometry is 15%. Background is said to be less than 3 cpm. Battery operated, the instrument utilizes high-resistance insulation in high-voltage circuits; guard rings are used throughout.

HIGH-VACUUM FURNACE

National Research Corp., 70 Memorial Drive, Cambridge, Mass. The type 29–0410–18 vacuum furnace operates at temperatures as high as 2 000° C and ultimate vacuums as low as 5×10^{-5} mm Hg. Power supply, vacuum pumping system, gages and controls are enclosed in a metal cabinet. The furnace bell jar is hinged to a vertical baseplate on top of the cabinet and is accessible from both sides. A single-turn low-voltage resistance element provides a constant temperature zone of 6 in. ×

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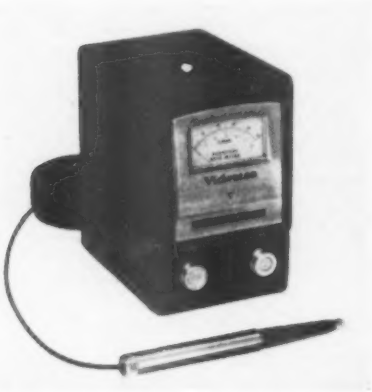
2½ in. diameter. Working temperature is reached in less than 1 min and is controllable within ±5° C. A thermocouple vacuum gage is standard equipment; other gages and special equipment for pouring and induction heating are available.



X-RAY RATE METER

Victoreen Instrument Co., 5806 Hough Ave., Cleveland 3, Ohio. The Roentgen Ratemeter consists of a thimble ionization chamber and preamplifier connected by a cable to a control unit containing a meter calibrated in r/min. Two probes are available: one gives full-scale meter readings of 3, 10, 30, and 100 r/min; the other, 30, 100, 300, and 1,000 r/min. Scale changing and zero-adjust controls are located on the control case.

MAGNETIC TRIGGER PAIR

Alden Products Co., 117 N. Main St., Brockton 64, Mass. Capable of handling pulses at rates as high as 25 kc, the model 5100RA Static Magnetic Memory can be used for storing, transferring, advancing and reading out information in computer circuits. Power is not required for storage of pulse information.

The unit consists of a three-winding miniature toroidal coil mounted on a card with a coupling circuit which facilitates two-core operation as a trigger pair. Current requirement for changing magnetization of the coil is about 125 ma. The unit is designed so that a pulse rise time of 5 μsec will give a minimum signal-to-noise ratio.

LITERATURE AVAILABLE

Tube Bending Chart. Slide-rule chart gives minimum bending radii for stainless-steel tubing, other pipe and tubing data in tabular form. *The Carpenter Steel Co., Alloy Tube Division, Union, N. J.*

Electromagnet. Brochure describes ADL electromagnet which provides variable pole-face area and air gap, field strengths as high as 35,000 gauss. *Arthur D. Little, Inc., 30 Memorial Drive, Cambridge 42, Mass.*

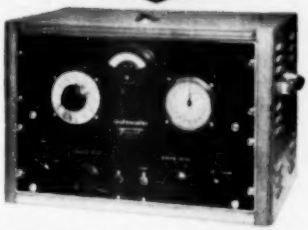
Valves. Pressure reducing and pressure relief valves are described in Bulletin 299. *A. W. Cash Valve Mfg. Corp., 666 E. Wabash Ave., Decatur, Ill.*

Power Supplies. Data sheets describe voltage-regulated power supplies with output voltages as high as 500 volts at currents as large as 300 ma. *Kepco Laboratories, Inc., 149-14 41st Ave., Flushing, N. Y.*

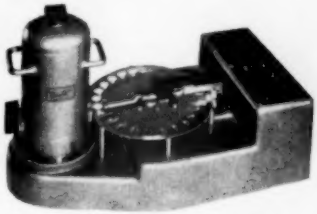
Rectifier Cartridges. Data sheet describes selenium-rectifier cartridges with voltage ratings up to 25 kv and current ratings to 75 ma. *International Rectifier Corp., 6809 S. Victoria Ave., Los Angeles 43, Calif.*

Synchronous Data Cameras. Bulletin details motion picture cameras designed for synchronous operation which permits time-basis comparison of data recorded at different points. *Flight Research Engineering Corp., P. O. Box 1-F, Richmond 1, Va.*

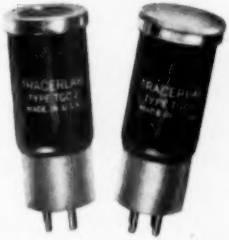
For
PRECISE RADIOASSAYS



AUTOSCALER — One of several scalers available for accurate radioactivity measurements.



AUTOMATIC SAMPLE CHANGER — For the automatic radioassay of large numbers of samples.



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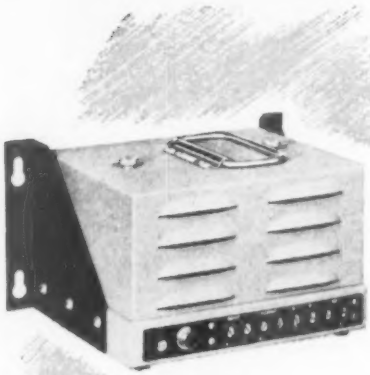

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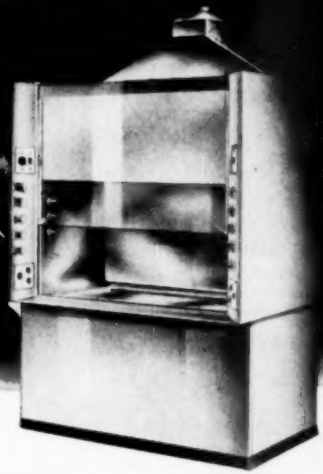

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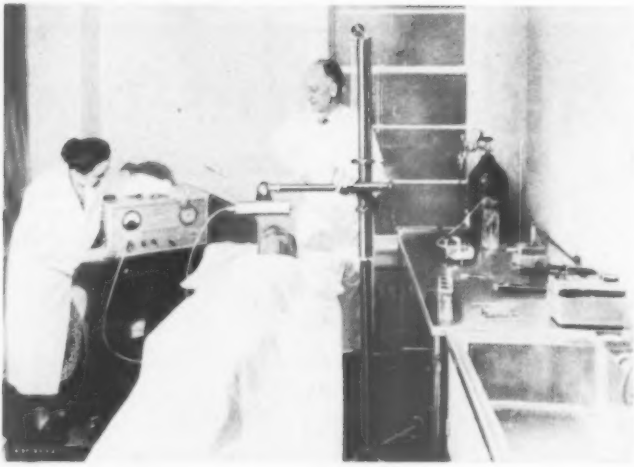

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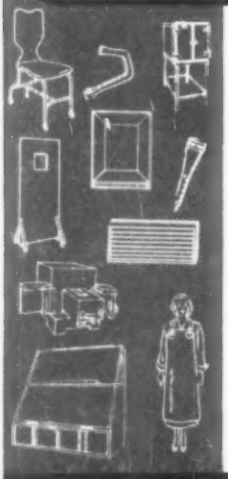

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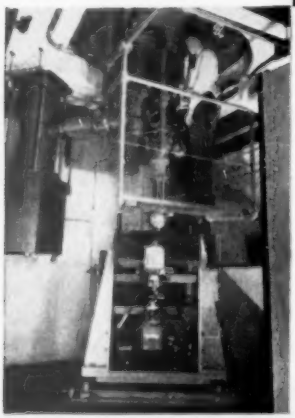

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intimate contact between the Atomic Energy Commission and industry, apart from the AEC contract operators. The best evidence of the general soundness of the Industrial Advisory Group view that once opportunities appear, industry will take advantage of them, are the industrial proposals now pending before the Commission for privately financed work on experimental reactors. But while these proposals are hopeful, much more ambitious private reactor work is desirable.

Magnitude of Stakes

This brings in a difficulty of truly formidable proportions, and one which I have termed as Magnitude of Stakes. Because of the large amounts of capital involved in starting any program of research, let alone a program of experimentation on a full scale in the atomic power field, things that are possible in other fields of economic endeavor such as in the chemical field, in the field of synthetic fibers, etc., are not possible in atomic power. In such other fields a large progressive industrial organization willing and able to stake a relatively few million dollars on research and development over a period of four to five years can bring itself to a position where, the research having proven successful, it can start and justify a full commercial scale operation by a capital investment, say of the order of 10 to 25 million dollars. Relatively large individual organizations in these fields can, therefore, undertake such programs on their own.

In the field of atomic power the required investment is so much higher, and the nature of the application is such, that even large organizations find it impossible to undertake on their own any such comparable programs. This is particularly so since the elements for evaluating the economic gain are still missing.

These then are some of the major obstacles to the participation of private industry in the development of atomic energy. It isn't possible, within the time and scope of this discussion, to go into the possible means or the mechanisms for removing these obstacles. I am, however, hopeful that such ways and means will be found and when that is done the path will be cleared for more effective participation of private industry—for the release of those creative forces that have brought us along so far on the path of scientific, technological and economic progress.

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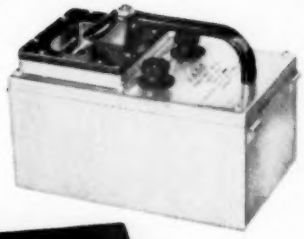

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nuclear instrument & chemical corporation
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December 29, 1950

[redacted]

Dear Doctor [redacted]

This is to tell you about some of our activities in the field of radioactive chemicals. Recently we began a program designed to furnish research workers with various chemicals synthesized with radioactive carbon. We plan to devote most of our attention to the preparation of substances by biological synthesis, with plants and with micro-organisms, to obtain radioactive chemicals that cannot be made in the synthetic organic chemistry laboratory.

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Very truly yours,

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John L. Kurans

J. L. Kurans
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